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# <sup>66</sup>Ga: a Standard for High-Energy Calibration of Ge Detectors

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#### Abstract

Two independent measurements of the relative emission probabilities for the strongest transitions in <sup>66</sup>Ga electron capture decay are reported here. The results of these measurements are in excellent agreement with each other and with those from another recent measurement. Consequently, <sup>66</sup>Ga emission probabilities for eighteen strong lines, from 834 keV to 4806 keV, are now known to better than 1% accuracy. Thus, <sup>66</sup>Ga can now be considered to be a suitable radionuclide for Ge detector efficiency calibration up to an energy of 4806 keV, the highest energy attainable with radioactive calibration sources. We have also provided an empirical function for correcting earlier incorrect emission probability results for  $E_{\gamma}$  greater than about 3 MeV which were produced using an inappropriate efficiency curve extrapolation.

Keywords: efficiency calibration, Ge detector, <sup>66</sup>Ga. PACS: 23.20.-g, 23.40.-s, 27.50.+e, 29.40.Wk. Corresponding author: C.M. Baglin, M/S 50A-1148, Lawrence Berkeley National Lab., 1 Cyclotron Rd., Berkeley, CA 94720, USA; Tel.: 1-408-779-4796; Fax: 1-510-486-5757; Email: baglin@lbl.gov

#### 1. Introduction

Due to the variation in the photoelectric, Compton, and pair-production cross sections for germanium as a function of the  $\gamma$ -ray energy, the response of a Ge detector is dependent not only on the specific detector geometry used but also on the  $\gamma$  ray energy. Consequently, one must calibrate the efficiency of a Ge detector throughout the energy range of interest. There are several commercially available radioactive sources that are commonly used for calibrating Ge detectors, such as <sup>22</sup>Na, <sup>24</sup>Na, <sup>54</sup>Mn, <sup>56</sup>Co, <sup>57</sup>Co, <sup>60</sup>Co, <sup>88</sup>Y, <sup>133</sup>Ba, <sup>137</sup>Cs, <sup>152</sup>Eu, <sup>154</sup>Eu, <sup>203</sup>Hg, <sup>207</sup>Bi, <sup>228</sup>Th and <sup>241</sup>Am and, for these, the  $\gamma$ -ray emission probabilities for the strongest lines are known with a precision of ≈1% or better [1]. Their energies, however, cover only the limited range of 14-3500 keV.

<sup>66</sup>Ga is potentially an important radionuclide for detector calibration in that, despite its relatively short half-life of 9.5 h, it can be produced readily through the <sup>66</sup>Zn(p,n) and <sup>63</sup>Cu( $\alpha$ ,n) reactions, and then used as a secondary standard to extend the calibration energy range to 4806 keV. Also, the  $\gamma$ -ray multiplicity of <sup>66</sup>Ga is low; consequently, any correction for coincidence summing will be small (provided that low-energy X-rays from the electron-capture process do not reach the detector). To use <sup>66</sup>Ga as a standard, however, its  $\gamma$ -ray emission probabilities must firstly be determined accurately, and it is here that a serious problem existed, as described below.

The relative γ-ray emission probabilities of <sup>66</sup>Ga were measured with good statistical precision in 1971 by Camp et al. [2] and a more extensive, but interrelated, measurement was reported in 1994 by Endt and Alderliesten [3] who normalized their emission probabilities for the strongest  $\gamma$  rays to the values deduced by Camp *et al.* [2]. Camp *et al.* determined their detector efficiency up to 2750 keV using radioactive sources, then assumed an almost *linear* extrapolation on a log-log plot of the efficiency curve between 1000 keV and 2000 keV to deduce the detector efficiency between 2500 keV and 5000 keV. It is now generally recognized that Ge detector efficiency curves do not exhibit the form assumed by Camp et al. in the 2500 to 5000 keV energy region. This conclusion was reached in 1974 by McCallum and Coote [4] who, having calibrated their detector efficiency up to 11,588 keV using  $\gamma$ -ray pairs from several (p, $\gamma$ ) resonances, measured emission probabilities for <sup>66</sup>Ga and <sup>56</sup>Co and observed that the values reported by Camp et al. [2] were systematically lower, by as much as 30% for the 4806-keV  $\gamma$  ray of <sup>66</sup>Ga. Also, they found that above 2500 keV their detector efficiency did not decrease linearly with energy on a log-log scale, but it decayed much more rapidly, indicating a lower capture rate in the detector for the higher-energy  $\gamma$  rays. Therefore, the emission probabilities for energies above about 2500 keV reported by Camp et al. [2] are systematically too low, a fact that has recently been corroborated by Schmid *et al.* [5].

McCallum and Coote [4] presented factors to correct the results from Camp *et al.* [2], and Trzaska [6] and Bhat [7] have taken these into account in their data evaluations when recommending  $\gamma$ -ray emission probabilities for <sup>66</sup>Ga. However, since the proposed correction factors lack explicit uncertainties, all of this effort produced new emission

probability values, but without associated experimental uncertainties; thus, <sup>66</sup>Ga remained unsuitable as a calibration standard. To remedy this situation, we undertook new emission probability measurements in two different laboratories (Berkeley and Budapest) using different detectors and calibration standards, as well as independently-prepared <sup>66</sup>Ga sources. It was only upon the completion of these measurements that we learned of the recent measurement of <sup>66</sup>Ga emission probabilities by Raman *et al.* [8]. In the present paper we give a detailed account of our measurements and present a recommended set of <sup>66</sup>Ga decay  $\gamma$ -ray emission probabilities useful for detector calibration.

## 2. Experimental Procedure

## 2.1 The Berkeley Measurement

## 2.1.1 66Ga Source Preparation

For this measurement, we produced  $^{66}Ga$  by bombarding a 1.6 mm thick foil of >99% pure natural zinc (48.6%  $^{64}Zn$ , 27.9%  $^{66}Zn$ , 4.1%  $^{67}Zn$ , 18.8%  $^{68}Zn$ , 0.62%  $^{70}Zn$ ) supplied by Alfa Products with a 17.5-MeV proton beam from the 88-Inch Cyclotron at the Lawrence Berkeley National Laboratory. At this beam energy,  $^{66}Ga$  is produced primarily via the  $^{66}Zn(p,n)$  reaction, with a small contribution from  $^{67}Zn(p,2n)$  [9].  $^{67}Ga(3.26 d)$  is also produced (via  $^{67}Zn(p,n)$  and  $^{68}Zn(p,2n)$ ), and a small amount of  $^{65}Zn(244.3 d)$  is expected following  $\beta$ - decay of  $^{65}Ga(15.2 min)$  produced by  $^{66}Zn(p,2n)$ ; the considerable  $^{68}Ga(67.6 min)$  activity produced by  $^{68}Zn(p,n)$  necessitated aging the source before use. After a cooling down period of about 26 hours, the source had a  $^{66}Ga$  activity of about 15  $\mu$ Ci. It was then placed in front of a shielded detector where we measured its gamma-ray spectrum

## 2.1.2 Detector

We used an Ortec 300 cm<sup>3</sup> (80% relative efficiency) p-type coaxial hyper-pure Ge detector without Compton suppression, connected to an ORTEC ACE data acquisition system operating on a PC. Spectra were recorded with the front face of the detector either 65 cm or 30.5 cm from the source. The long-term energy resolution (FWHM) obtained ranged from approximately 1.8 keV at 800 keV to 3.7 keV at 4800 keV.

## 2.1.3 Detector Efficiency Calibration

Since the precision in the gamma-ray detector's efficiency determination as a function of energy would ultimately contribute to our uncertainties in the emission probabilities of the various gamma rays from <sup>66</sup>Ga, it was of the utmost importance to calibrate the detector carefully and precisely. The *absolute* full energy peak efficiency for a given energy and geometry,  $\varepsilon(E_{\gamma})$ , is given by:

$$\varepsilon(E_{\gamma}) = N_{\gamma} / (AI_{\gamma}) . \tag{1}$$

Here,  $N_{\gamma}$  is the count rate of a particular  $\gamma$  ray in the spectrum, A is the source activity, and  $I_{\gamma}$  is the absolute  $\gamma$ -ray emission probability.

For the *relative* efficiency calibration, we measured  $\gamma$ -ray intensities on an arbitrary scale for the well-known multi  $\gamma$ -ray standards <sup>56</sup>Co, <sup>152</sup>Eu, <sup>154</sup>Eu and <sup>228</sup>Th. To determine *absolute* efficiency values, we used sources of <sup>60</sup>Co and <sup>137</sup>Cs (obtained from Isotopes Products Laboratories) whose activities were known to ±1% relative standard uncertainty and a <sup>13</sup>C(<sup>238</sup>Pu) source (from Laboratoire de Métrologie des Rayonnements Ionisants, France) whose combined source strength plus absolute emission probability was known to an accuracy of ±6%. The latter source's 6129-keV calibration  $\gamma$  ray, produced via the <sup>13</sup>C( $\alpha$ ,n)<sup>16</sup>O reaction in the <sup>13</sup>C(<sup>238</sup>Pu) source, provided a high-energy point which was essential for achieving a reliable fit of the calibration curve through the 3500-6000 keV energy range. In contrast to the 1% and 6% accuracies of the source strengths used for absolute efficiency calibration, most of the relative  $\gamma$ -ray emission probabilities used for the relative efficiency calibration have uncertainties of 1% or less. For these, we used the values recommended in the IAEA-TECDOC-619 report [1], except for <sup>56</sup>Co, which has high-energy  $\gamma$  rays that required further evaluation, as described in the Appendix.

#### 2.1.4 Data Analysis Software

Hypermet-PC [10], a commercial software package developed at Budapest on the basis of the well-known HYPERMET code [11], provided the routines both to analyze  $\gamma$ -ray spectra and to calculate detector efficiencies and fit an analytical function to them. The program fits a Gaussian function with an exponential low-energy tail to each peak in the automatically selected range in the spectrum, and a second-order polynomial combined with two kinds of step functions to the underlying background. It calculates and displays on the screen the peak area (count rate), the  $\chi^2/\nu$  value, and the residuals of the fit for each fitted region. The program also calculates the detector efficiency using the spectral data and the recommended  $\gamma$ -ray emission probabilities provided by the user. Since the detector efficiency is a continuous function of the  $\gamma$ -ray energy, the relative precision of the interpolated values may be somewhat higher than that of the nearby experimental points. Thus it is feasible to measure relative emission probabilities with a precision better than that of the individual experimental efficiencies. Hypermet-PC fits a polynomial function to the logarithms of the experimental efficiencies and allows the user to choose the polynomial order. The program interpolates efficiency values for relevant  $\gamma$ -ray energies using this polynomial function and applies them to correct the spectral peak areas (count rates) for the detector response at different energies. The reliability and accuracy of this procedure are demonstrated in a parallel paper [12].

For our detector, fitting with a second-order polynomial in a limited range of  $\gamma$ -ray energies (661-6129 keV) was preferable to using a higher-order polynomial for a wider energy range (121-6129 keV). This approach avoids non-physical oscillations at either extreme of the energy range. Forty-five experimental efficiency values were fitted with  $\chi^2/\nu = 0.95$ .

Figure 1 shows the efficiency curve for the Berkeley Ge detector as well as the residuals of the fit, given as experimental value minus fitted value divided by the experimental uncertainty. Notice that the latter have a dispersion of  $\approx$ 2 standard deviations about a central value of zero, which gives an indication of the general precision with which the <sup>66</sup>Ga relative  $\gamma$ -ray emission probabilities have been determined here. The program simultaneously fitted the relative efficiencies for  $\gamma$ -rays from each of the standards to produce a single curve for the entire energy range. The certified activity data for <sup>137</sup>Cs, <sup>60</sup>Co and <sup>13</sup>C(<sup>238</sup>Pu) have been included to normalize this curve to an absolute scale (per  $\gamma$  ray emitted for a given geometry) whereas, for the rest of the data sets, freely variable normalizing constants have been introduced in the fitting procedure. Clearly, a linear extrapolation (on a log-log plot) beyond 2000 keV would, for this detector, result in an efficiency which is much too high by 5000 keV. It was this type of extrapolation by Camp *et al.* [2] which led to incorrect values for the relative emission probabilities of  $\gamma$  rays from <sup>66</sup>Ga decay in the 3000-5000 keV energy range.

#### 2.1.5 66Ga γ-ray Spectrum

We initially placed our <sup>66</sup>Ga source 65 cm away from the detector face, and recorded on magnetic disk nine 8192-channel  $\gamma$ -ray spectra, each of two hours duration. These enabled a rough verification that the count rates of several of the strongest peaks decayed with a half-life of about 9 h as expected for <sup>66</sup>Ga. Another <sup>66</sup>Ga-source spectrum was subsequently recorded with a 30.5 cm separation between source and detector. Comparison of the <sup>66</sup>Ga spectra recorded for different source to detector distance, source strength and source age enabled a valuable check on peak identification and on possible summing effects. Because of its superior statistics, however, only the summed spectrum from the 65 cm measurement was ultimately used to deduce the relative emission probabilities. The  $\gamma$  rays from <sup>67</sup>Ga, produced via the <sup>67</sup>Zn(p,n) and <sup>68</sup>Zn(p,2n) reactions, were the only significant source impurity lines observed (see Figure 2).

We also measured a background spectrum following these measurements. This spectrum revealed the existence of several  $\gamma$  rays, among them those at 1173 keV and 1332 keV (<sup>60</sup>Co), 1275 keV (<sup>22</sup>Na), 1461 keV (<sup>40</sup>K), 1766 keV (<sup>226</sup>Ra - <sup>214</sup>Bi decay chain), and 2615 keV (<sup>232</sup>Th - <sup>208</sup>Tl decay chain). Most of these were too weak to be observed in our <sup>66</sup>Ga spectrum, and did not interfere with its peaks. However, we oberved a weak 1173-keV peak in the <sup>66</sup>Ga spectrum, so the contribution from the 1332-keV  $\gamma$  ray (also from <sup>60</sup>Co) had to be removed from the 1333-keV peak from <sup>66</sup>Ga. We applied a 1.8% correction to this peak area, based on the magnitude of the count rate of the 1173-keV impurity  $\gamma$  ray in the <sup>66</sup>Ga spectrum.

A much more significant adjustment to the count rate of the 4295-keV  $\gamma$  ray was required, originating from the superposition of its full-energy peak on the single-escape peak of the 4806-keV  $\gamma$  ray. This correction required knowledge of the single-escape to photopeak ratio for a 4806-keV  $\gamma$  ray in our detector. Figure 3 shows experimental values of this ratio as a function of the  $\gamma$ -ray energy, deduced from various spectra recorded with the same

geometry. An interpolated value of 0.440 (10) at 4806 keV led to a 16% adjustment in the count rate of the 4295-keV peak.

The uncertainties in our measured emission probabilities range from 1% to 2%, and include contributions from both the spectral statistics and the peak shape analysis (less than about 1% for most transitions) and from the detector efficiency (0.8% to 1.2%).

#### *2.2 The Budapest Measurement* 2.2.1 <sup>66</sup>Ga Source Preparation

Enriched <sup>66</sup>Zn targets were prepared by electrodeposition. The isotopic composition of the <sup>66</sup>Zn was <sup>64</sup>Zn (0.4%), <sup>66</sup>Zn (99.0±0.1%), <sup>67</sup>Zn(0.17%), <sup>68</sup>Zn(0.43%) and <sup>70</sup>Zn(0.03%). The material was supplied by v/o Techsnabexport, Moscow, Russia. The electrolyte was 50 ml 0.5 M HCl containing 0.5 g enriched <sup>66</sup>ZnCl<sub>2</sub>. The time of electrolysis was 3 minutes at a constant current of 200 mA. Targets of about 0.015 g/cm<sup>2</sup> thickness were electrodeposited onto 5-µm thick natural nickel foils (from Goodfellow Metals, Cambridge, UK). Natural Zn targets of 10 µm thickness were also obtained from Goodfellow Metals. The diameter of the natural Zn and Ni foils was 2 cm, whereas the enriched targets had a diameter of 1 cm.

Irradiation was performed with the external beam of the MGC 20E cyclotron of ATOMKI Debrecen, Hungary. The energy of the extracted proton beam was 14.5 MeV. Five stacked targets (three of enriched <sup>66</sup>Zn and two of natural Zn) were activated. Direct measurement of beam intensity was carried out with a Faraday cup that served also as a target holder. The average beam current was 150 nA and the irradiation time was 6.5 h. The diameter of the beam was 6 mm. The total <sup>66</sup>Ga activity from the five foils together was about 5.5 mCi at EOB (End of Bombardment), hence the targets had to be cooled for one day before starting the measurements.

## 2.2.2 Detector

The experiments were performed at the capture  $\gamma$ -ray facility [13,14] of the Budapest Research Reactor. The  $\gamma$ -ray spectrometer consists of a Canberra n-type, high-purity germanium main detector with closed-end coaxial geometry (25% efficiency, about 100 cm<sup>3</sup> active volume and 1.8 keV resolution at 1332 keV) and a Bicron BGO scintillator guard detector annulus, surrounded by a 10 cm thick lead shield. The Compton-suppression enhancement is about 5 at 1332 keV, and reaches 40 at about 7 MeV. Single- and double-escape peaks are suppressed by a factor of 10 and 100, respectively. The data were collected by a personal computer-based MCA, with 16k conversion gain. The dispersion was set to 0.61 keV per channel to encompass the broad energy range covered by the efficiency calibration lines which included neutron-capture  $\gamma$  rays.

The measurements were performed in Compton-suppressed mode, at source to detector window separations of 23.5 cm (standard position) and 31.5 cm. The diameter of the sources and capture targets used for efficiency calibration did not exceed 0.5 cm, whereas the  $\gamma$ -ray collimator aperture was 2 cm. The long-term energy resolution (FWHM) was about 1.9 keV at 1 MeV, and 3.7 keV at 4.8 MeV.

## 2.2.3 Detector Calibration and Data Analysis

Energy and efficiency calibrations were performed using <sup>133</sup>Ba and <sup>152</sup>Eu multi-gamma radioactive sources, recommended in the IAEA-TECDOC-619 [1] as secondary standards, as well as neutron capture gamma rays from the <sup>35</sup>Cl(n, $\gamma$ ) reaction, measured previously at Budapest [15] with respect to the <sup>14</sup>N(n, $\gamma$ ) reaction as a primary standard [1].

The spectra were fitted using Hypermet-PC [10], which also includes routines for efficiency and non-linearity calibrations. The procedures for spectrum fitting and efficiency calibration have been described in detail in sections 2.1.3-2.1.4. Although the <sup>152</sup>Eu source was a calibrated standard, its certified activity was used to set a realistic scale for the fitted relative efficiency only. Therefore, no additional uncertainties have been included beside the statistical uncertainties and the uncertainties in the data from the literature.

The relative full-energy peak efficiency has been determined for the entire energy range spanned by the calibration sources used, i.e., from 81 keV to 8576 keV. The 54 experimental values could be fitted by a seventh-order polynomial, with  $\chi^2/\nu = 0.78$  and 0.91 for the 23.5 cm and 31.5 cm distances, respectively. Figure 4 illustrates the excellent fit and the small uncertainties of the fitted efficiency function (less than about 0.6% throughout the energy range of interest here).

# 2.2.4 66Ga γ-ray Spectrum

The irradiation of three targets enriched in <sup>66</sup>Zn and two targets of natural Zn provided three 'enriched' and two 'natural' <sup>66</sup>Ga sources. Hence it was possible to keep the counting rate of the Compton-suppressed spectrum relatively stable and below 1 kHz by successively adding more sources and by changing the source-to-detector distance. The <sup>66</sup>Ga lines spanned the first 8k of the entire 16k spectrum. Eventually, two cumulative spectra, measured for about 20 h at each distance with enriched sources, and another one measured for 2.5 h with a natural source at 23.5 cm provided the best data.

Figure 5 shows a typical <sup>66</sup>Ga decay spectrum, recorded with an enriched source 23.5 cm away from the detector. Due to the relatively long irradiation time, the <sup>67</sup>Ga peaks appear despite the low abundance of <sup>67</sup>Zn in the target. Moreover, <sup>55</sup>Co (17.5 h), <sup>57</sup>Co (272 d) and <sup>57</sup>Ni (35.6 h) have been produced by the irradiation of the thin Ni target backing. From this viewpoint, the use of a self-supporting natural Zn target would have been more

advantageous. It is also interesting to note that  ${}^{65}$ Zn, the long-lived decay product of 15.2min  ${}^{65}$ Ga produced in the  ${}^{66}$ Zn(p,2n) reaction was observed (see the strong 1115-keV line). To suppress it, one should use a somewhat lower proton beam energy [9]. Fortunately, none of the strong peaks from  ${}^{66}$ Ga was seriously affected by the aforementioned contaminants.

The Compton suppression greatly reduces the intensity of the escape peaks. Double-escape peaks were practically eliminated, and only the single-escape peak of the strong 4806-keV  $\gamma$  ray interferes with a <sup>66</sup>Ga peak of interest. The area of the 4295-keV peak had to be corrected for a 6% contribution from this single-escape peak. On the other hand, due to the uncovered solid angle in the backward direction, the Compton edges look like broad peaks, as seen in Figure 5, complicating the peak fitting. For instance, the 834-keV peak is sitting on the right-hand side of one such "peak".

## 3. Discussion of Results

Table 1 shows the <sup>66</sup>Ga relative  $\gamma$ -ray emission probabilities determined in the present measurements, along with those those from Camp. *et al.* [2] and from Raman *et al.* [8]. We report here only those  $\gamma$  rays with emission probabilities greater than 1% of that of the 1039-keV  $\gamma$  ray, since these are the most useful for Ge-detector calibration. The newly recommended  $\gamma$ -ray energy values from Helmer and van der Leun [16] have been given to identify the lines.

In Figure 6a we have plotted the ratio of the emission probabilities determined in the Berkeley measurement to those from the Budapest measurement. These two sets of data are in excellent agreement. All ratios agree within two standard deviations, and most of them within 2.5%. Figure 6b shows the ratios of the weighted averages of the present Berkeley and Budapest emission probabilities to those from Raman *et al.* [8]. The agreement is excellent. Hence, we recommend that the weighted average of the values from all three measurements be used for detector efficiency calibration. These are summarized in the right-hand column of Table 1.

Figure 7 is a plot of our recommended emission probabilities (from Table 1) divided by those from Camp *et al.* [2]. Here we see a systematic deviation from a ratio of 1.00. The Camp *et al.* data are several percent too high near 2000 keV, but more than 5% too low at 3500 keV and about 30% too low at 4806 keV. Moreover, the 4295-keV point is out of trend, probably due to an improper correction for the 4806-keV single escape contribution in their work. We have also plotted in this figure the correction factors for the emission probabilities of Camp *et al.* [2] that were proposed by McCallum and Coote [4]. Note that, although the correcting formula of McCallum and Coote [4] did not include uncertainties because "systematic errors of unknown magnitude are likely to be involved" [4], those authors estimated a statistical precision of about 5% which, from Figure 7, appears to be entirely realistic.

For those who used the intensity data from Camp *et al.* [2] for detector calibration and now wish to correct their old data, it may be useful to have the new correction function (by which the  $E_{\gamma}>1$  MeV data of Camp *et al.* should be multiplied) in its analytical form, namely:

$$f(E_{\gamma}) = 1.116 (11) - 0.155 (11) E_{\gamma} + 0.0397 (22) E_{\gamma}^2$$
. (2)

 $E_{\gamma}$  is the  $\gamma$ -ray energy in units of MeV.  $\chi^2/\nu = 0.41$  is the reduced chi-squared for the second-order polynomial fit. The uncertainty (in the least-significant digits) in each coefficient (calculated from the relevant diagonal element of the covariance matrix) is given in parentheses.

#### 4. Conclusions

The present two independent measurements of the <sup>66</sup>Ga relative  $\gamma$ -ray emission probabilities performed at Berkeley and Budapest have provided precise values that agree well with each other. A new set of data from Raman *et al.* [8] is also in excellent agreement with our results. Therefore, we conclude that <sup>66</sup>Ga is now a radionuclide that may reliably be used for calibration of Ge detectors up to 4806 keV. We recommend the weighted average of the results from these three measurements (Table 1) for use as a detector calibration standard until such time as a new critical data evaluation for <sup>66</sup>Ga emission probabilities becomes available.

These results also reaffirm the need for a global revision of all emission probability data for  $E_{\gamma}$  greater than about 2500 keV which have been determined using detectors whose calibration depended on the <sup>56</sup>Co or <sup>66</sup>Ga relative emission probabilities from Camp *et al.* [2], or on the assumption of a linear extrapolation of efficiency versus energy on a log-log scale. Obvious examples are the <sup>56</sup>Co measurements of Hofmann [27] and the extensive <sup>66</sup>Ga measurements by Endt and Alderliesten [3]. To facilitate recalibration of results depending on the Camp *et al.* data, we have provided a new correction formula as well.

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#### Appendix: <sup>56</sup>Co γ-ray Emission Probabilities

Among the commonly used radionuclide standards for efficiency calibration of Ge detectors, <sup>56</sup>Co is the one that emits  $\gamma$  rays with the highest energies (up to about 3500 keV). Because <sup>66</sup>Ga emits  $\gamma$  rays with energies up to 4806 keV, it was particularly important to have accurate values for the emission probabilities of <sup>56</sup>Co in order to provide detector efficiency calibration points in the 2500-3500 keV energy range. Since the IAEA-TECDOC-619 [1] evaluation for <sup>56</sup>Co did not include some important measurements, we performed our own evaluation of the available data.

We summarize in Table 2 the relative emission probabilities from various measurements in the literature. We separated the measurements into two groups according to the methods used to determine the detector efficiencies at energies greater than about 2600 keV. The first group comprises studies for which those efficiencies were determined by using  $(n, \gamma)$  reactions or  $(p, \gamma)$  resonances (Boydell [18], Gehrke [20], Hautala [21], Meyer [17]) or by using <sup>88</sup>Rb (17.8 min) (to 3218 keV) and <sup>49</sup>Ca (8.7 min) (to 4072 keV) radioactive sources (Stewart [22]). The other group is made up of studies which relied either on *linear* extrapolations of the efficiency on a log-log plot (Camp *et al.* [2]), or on Monte Carlo calculations (Schötzig et al. [24]). We consider less reliable (probably affected by systematic errors) the emission probabilities from this second group. Consequently, we included in our averages the data from all independent measurements except that of Schötzig et al. [24] for energies below 2598 keV, but only values from the first group above this energy. All data from Schötzig et al. were omitted because the calibration sources they used, and hence the extent to which their calibration relied upon Monte Carlo calculations, was not known. A measurement by Phelps et al. [26] was rejected also because the detector calibration for that experiment included earlier <sup>56</sup>Co data from Camp et al. [2], almost certainly introducing an interdependence between data from Phelps et al. [26] and from Camp *et al.* [2].

The newly-evaluated values for <sup>56</sup>Co relative emission probabilities are given in Table 2, in the second to last column. Except for the weakest lines (as noted in Table 2), these were used as part of the Berkeley detector efficiency calibration. They are weighted averages of the measurements mentioned above unless those measurements were discrepant. The latter is considered to be the case when their  $\chi^2/\nu$  exceeds a critical value which depends on the number of degrees of freedom [28]; the average of such data has been determined using the Method of Limitation of Relative Statistical Weight [25]. This method also uses a weighted average, but it imposes a requirement that no single datum in the set of discrepant data should contribute a relative statistical weight greater than 50%. Several values listed in Table 2 are statistical outliers according to the Chauvenet criterion [29]; these have been appropriately labelled. Values of  $\chi^2/\nu$  which exceed the critical value are also identified. The emission probabilities are given relative to a value of 100 for the 847keV transition. When the authors gave no uncertainty in a datum, we conservatively assigned a relative uncertainty equal to that for their measurement of the next-strongest transition (i.e., that at 1238 keV). While we were writing this paper, two independent,

high-precision determinations of relative transition probabilities for  ${}^{56}$ Co became available, one by Molnár *et al.* [12,15], the other by Raman *et al.* [8]. Both are in excellent agreement with the average values presented in Table 2, as shown elsewhere [12,15].

# **Figure Captions**

#### Figure 1.

Absolute full-energy peak efficiency (left-hand scale) as a function of  $\gamma$ -ray energy for the Berkeley detector. The residuals of the fit to the data points (experimental value minus fitted value divided by the experimental uncertainty) are shown at the top of the figure. The dashed curve represents the relative standard uncertainties for the fitted curve (in percent, right-hand scale).

## Figure 2.

 $^{66}$ Ga  $\gamma$ -ray spectrum obtained using the Berkeley detector. The strongest  $^{66}$ Ga photopeaks are labeled with their energy (rounded to the nearest keV) and  $^{66}$ Ga escape peaks are designated by the symbol '\*'. Peaks labeled '&' are  $^{67}$ Ga photopeaks arising from the  $^{67}$ Zn and  $^{68}$ Zn components of the natural Zn target, and those labeled 'B' are room background photopeaks.

#### Figure 3.

Plot of the ratio of single-escape peak area to full-energy peak area as a function of the full-energy peak energy for the Berkeley detector.

#### Figure 4.

Absolute full-energy peak efficiency ( left-hand scale) as a function of  $\gamma$ -ray energy for the Budapest detector. The residuals of the fit to the data points (experimental value minus fitted value divided by the experimental uncertainty) are shown at the top of the figure. The dashed curve represents the relative standard uncertainties for the fitted curve (in percent, right-hand scale).

## Figure 5.

<sup>66</sup>Ga γ-ray spectrum of the enriched source obtained using the Compton-suppressed Budapest detector. The strong <sup>66</sup>Ga photopeaks are labeled with their energy (rounded to the nearest keV) and <sup>66</sup>Ga escape peaks are designated by the symbol '\*'. The <sup>67</sup>Ga photopeaks arising from a source impurity are labeled as such, while those from the isotopes produced in the Ni backing are marked by triangles. Compton edges are marked by 'C'.

#### Figure 6.

Comparison of measured <sup>66</sup>Ga emission probabilities as a function of  $\gamma$ -ray energy: (a) ratios of Berkeley data to Budapest data from the two present experiments; (b) the weighted average emission probability determined from the present two measurements divided by the emission probability determined by Raman *et al.* [8].

#### Figure 7.

The relative  $\gamma$ -ray intensities recommended in Table 1 divided by the intensities from Camp *et al.* [2]. The solid curve is a fit according to Eq. 2. The dashed curve represents

the correction factor  $F=1.053-0.079E_{\gamma}+0.026E_{\gamma}^2$  (where  $E_{\gamma}$  is in MeV), proposed by McCallum and Coote [4] to correct the emission probabilities from Camp *et al.* [2]. The dashed-dotted line at the bottom of the figure represents the relative standard uncertainties for the new correction function (see right-hand scale).

E <sub>γ</sub> (keV) Helmer [16]	I <sub>γ</sub> (rel.) Camp <i>et al</i> . [2]	I <sub>γ</sub> (rel.) Raman <i>et al</i> .[8]	I <sub>γ</sub> (rel.) Berkeley	$I_{\gamma}$ (rel.) Budapest	$I_{\gamma}$ (rel.) Recommended
833.5324 (21)	15.92 (17)	16.02 (24)	15.94 (14)	15.92 (6)	15.93 (5)
1039.220 (3)	100.0	100.0 (16)	100.0 (9)	100.0 (3)	100.0 (3)
1333.112 (5)	3.25 (4)	3.17 (5)	3.20 (3)#	3.171 (13)	3.175 (12)
1418.754 (5)	1.70 (3)		1.640 (23)	1.659 (8)	1.657 (8)
1508.158 (7)	1.520 (24)		1.503 (23)	1.496 (7)	1.497 (7)
1898.823 (8)	1.09 (4)		1.062 (23)	1.050 (8)	1.051 (8)
1918.329 (5)	5.63 (8)	5.33 (8)	5.44 (6)	5.360 (23)**	5.368 (21)
2189.616 (6)	15.06 (18)	14.54 (21)	14.50 (13)	14.39 (6)	14.42 (5)
2422.525 (7)	5.16 (5)	5.12 (8)	5.15 (6)	5.072 (24)	5.085 (22)
2751.835 (5)	61.2 (6)	61.2 (8)	61.5 (6)	61.34 (26)	61.35 (23)
3228.800 (6)	3.96 (4)	4.06 (8)	4.07 (4)	4.087 (22)	4.082 (19)
3380.850 (6)	3.78 (4)	3.96 (8)	3.99 (4)	3.950 (23)	3.960 (19)
3422.040 (8)	2.18 (4)		2.29 (3)	2.321 (16)	2.314 (14)
3791.036 (8)##	2.68 (3)	2.96 (5)	2.96 (4)	2.929 (24)	2.941 (19)
4085.853 (9)	3.07 (4)	3.38 (8)	3.42 (4)	3.455 (20)	3.445 (18)
4295.224 (10)##	9.17 (11)	10.24 (26)*	10.54 (15)&	10.25 (7)@	10.30 (8) <sup>a</sup>
4461.202 (9)	1.875 (22)		2.20 (4)	2.275 (23)	2.26 (3)
4806.007 (9)	3.82 (4)	4.93 (11)	5.00 (7)	5.04 (3)	5.03 (3)

Table 1. Comparison of Measured Relative γ-Ray Emission Probabilities for 66Ga

 $^{\rm \#}$  Corrected for 1.8% background contribution from  $^{60}Co.$ 

& Corrected for 16% contribution from single-escape peak from 4806-keV  $\gamma$  ray.

\* Corrected for ~4% contribution from single-escape peak from 4806-keV  $\gamma$  ray.

\*\* Corrected for up to 4% contribution from <sup>57</sup>Ni for enriched sources.

<sup>@</sup> Corrected for 6.1(4)% contribution from single-escape peak from 4806-keV  $\gamma$  ray. <sup>##</sup> From fit to level scheme; not recommended for energy calibration [16]. <sup>a</sup> After correction for single-escape contribution from the 4806 keV line.

#### Table 2. <sup>56</sup>Co Relative γ-Ray Emission Probabilities

Energy (keV)	I <sub>r</sub> (rel) [20] Gehrke	I₁(rel) [21] Hautala	I <sub>/</sub> (rel) [22] Stewart	I <sub>γ</sub> (rel) [18] Boydell	I <sub>γ</sub> (rel) [2] Camp	I <sub>r</sub> (rel) [19] Katou	I <sub>Y</sub> (rel) [23] Yoshizawa		Average I <sub>γ</sub> (rel)	$\chi^2/N$
847	100.0 (10)	100.0 (6)	100.0 (13)	100.0 (15)	100.0 (10)	100.0 (11)	100.0 (3)	100.0 (7)	100.0 (2)	
977	1.426 (15)	1.38 (4)	1.41 (2)		1.448 (14)	1.386 (15)	1.435 (16)	1.440 (15)	1.425 (9)&	2.1
1038	14.04 (14)	13.5 (2)*	14.11 (19)	13.7 (6)	14.24 (14)	13.92 (12)	14.16 (5)	14.0 (1)	14.09 (5)	2.2
1175	2.28 (2)	2.11 (10)*	2.300 (32)	2.3 (1)	2.300 (25)	2.180 (24)	2.241 (12)	2.28 (2)	2.255 (14)	3.1#
1238	66.4 (7)	65.1 (4)	68.5 (9)	66.2 (10)	67.6 (7)	66.4 (7)	66.06 (21)	67.6 (4)	66.3 (3)	4.3#
1360	4.24 (4)	4.24 (15)	4.32 (6)	4.4 (1)	4.34 (5)	4.19 (5)	4.265 (17)	4.33 (4)	4.274 (15)	1.4
1771	15.65 (16)	15.26 (15)	15.5 (4)	15.9 (3)	15.78 (16)	15.37 (24)	15.49 (5)	15.70 (15)	15.52 (5)	1.4
1811	0.650 (7)	0.59 (3)*	0.629 (13)		0.641 (8)	0.665 (23)	0.657 (23)	0.640 (10)	0.643 (4)&	1.1
1964	0.724 (8)	0.70 (2)	0.719 (15)		0.721 (15)	0.667 (21)*	0.707 (11)	0.720 (15)	0.715 (6)&	1.3
2015	3.09 (5)	2.97 (3)	3.18 (7)	3.1 (1)	3.095 (31)	3.03 (7)	3.026 (14)	3.08 (3)	3.041 (16)	2.4
2035	7.95 (12)	7.64 (6)	8.14 (17)	7.8 (1)	7.95 (8)	7.69 (15)	7.766 (28)	7.89 (7)	7.79 (4)	2.8#
2113	0.387 (8)	0.34 (2)	0.375 (14)		0.387 (4)	0.375 (17)	0.363 (7)	0.350 (10)	0.378 (9)&	3.7#
2213	0.406 (9)	0.39 (2)	0.42 (2)*		0.377 (10)	0.387 (18)	0.389 (8)	0.385 (5)	0.389 (4)&	1.3
2598	17.34 (26)	17.19 (15)	17.4 (4)	17.3 (4)	16.85 (17)	16.64 (22)	16.96 (6)	17.29 (15)	17.02 (6)	1.8
3010	1.06 (3)	1.05 (3)	0.84 (4)*	1.0 (2)				1.05 (1)	1.03 (3)&	<b>6</b> .3 <sup>#</sup>
3202	3.18 (10)	3.24 (3)	3.03 (7)*	3.2 (1)				3.24 (3)	3.22 (3)	2.1
3253	7.79 (24)	7.97 (11)	7.60 (15)	8.2 (4)				7.94 (7)	7.90 (6)	1.4
3273	1.85 (6)	1.84 (3)	1.815 (36)	1.9 (1)				1.89 (2)	1.864 (15)	1.1
3451	0.93 (3)	0.95 (2)	0.90 (2)	1.00 (4)				0.954 (10)	0.945 (11)	2.0
3548	0.190 (6)	0.196 (5)	0.196 (6)	0.20 (2)				0.198 (5)	0.195 (3)	0.29
3601	0.0165 (7)	0.012 (3)	0.015 (2)					0.0180 (10)	0.0167 (7)&	1.7
3612	0.0085 (4)	0.005 (2)	0.010 (2)						0.0084 (5)&	1.8

\* This datum is a statistical outlier based on the Chauvenet criterion [29].

#  $\chi^2 N$  exceeds critical value, identifying these data as discrepant.

& Not utilized for Berkeley detector efficiency calibration.













