8

True Coincidence Summing

8.1 INTRODUCTION

In Chapter 7, I referred, without going into any detail, to the difficulties caused by true coincidence summing in achieving a valid efficiency calibration for close geometry measurements. The problem is not a new one (although as larger detectors become available, it becomes more significant) but appears often to be ignored in practice. The problem is important enough, and has been neglected long enough, to devote a chapter to it alone.

The problems caused by **true coincidence summing** (TCS) can be demonstrated by referring to the calibration curves for a 45 % HPGe detector shown in Figure 8.1. These were derived using ^{152}Eu near point sources of moderate activity such that the count rate in each case was about 7700 cps. The lower curve was measured with the source 115 mm from the detector, is smooth, consistent, and is, apparently, satisfactory.

The upper curve, measured with the source on the detector cap, is not at all satisfactory. The points do not lie on an orderly line and it would be difficult to draw an acceptable curve through them. The reason for this dramatic difference is TCS.

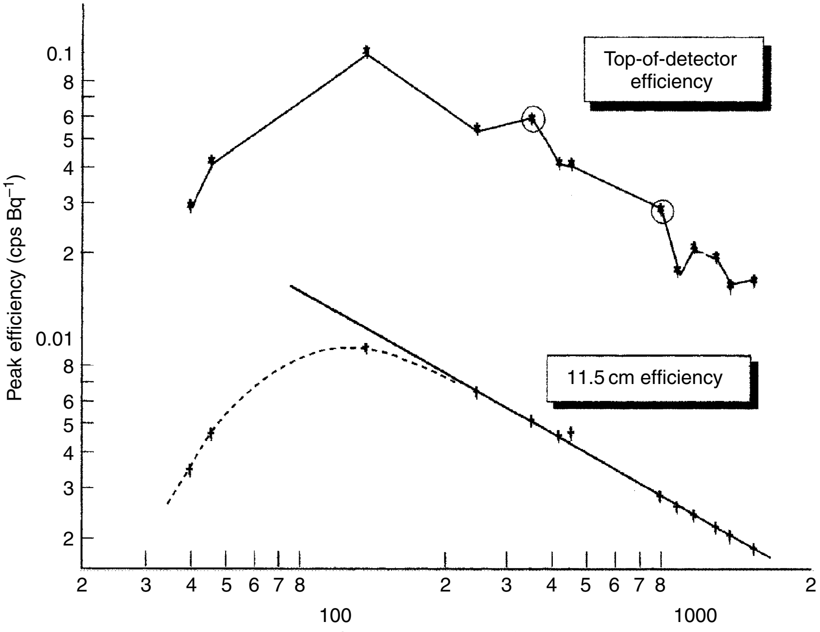

Figure 8.1 Efficiency curves on top of and at 115 mm from the detector endcap using ^{152}Eu

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8.2 THE ORIGIN OF SUMMING

Figure 8.2 shows a simplified decay scheme for ^{152}Eu. Atoms of this nuclide have a choice when they decay; they can emit a β^- particle and become ^{152}Gd or, more likely (on 72.08 % of occasions), undergo electron capture and become ^{152}Sm. Whatever the mode of decay, the daughter nucleus then de-excites by emitting a number of gamma-rays in one or other of the decay schemes. We must also remember that every electron capture decay to ^{152}Sm is likely to be accompanied by the emission of Sm X-rays.

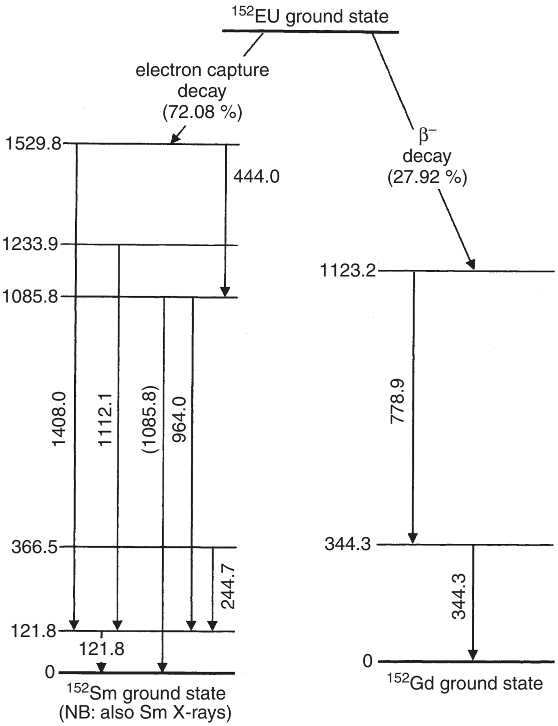

Figure 8.2 Simplified decay scheme for ^{152}Eu

The lifetimes of the individual nuclear levels are short, much shorter than the resolving time of the gamma-spectrometer system. From the point of view of the detector, every disintegration of a ^{152}Eu atom in the source will release a number of gamma-rays, and possibly X-rays, simultaneously and there is a certain probability that more than one of these will be detected together. If this happens, then a pulse will be recorded which represents the sum of the energies of the two individual photons. This is true coincidence summing; sometimes called **cascade summing**. It is the summing of two gamma-rays, or a gamma-ray and an X-ray, emitted in coincidence.

As with random summing, the event results in loss of counts from the full-energy gamma-ray peaks and a loss of efficiency. However, unlike the random summing that I discussed in Section 4.8, the summed pulse will not be misshapen and cannot be rejected by pile-up rejection circuitry.

8.3 SUMMING AND SOLID ANGLE

The degree of TCS depends upon the probability that two gamma-rays emitted simultaneously will be detected simultaneously. This is a function of geometry, of the solid angle subtended at the detector by the source. Figure 8.3 illustrates the geometrical arrangement under which the calibration curves in Figure 8.1 were measured. With the source on the detector cap, there is a 42 % chance that any gamma-ray will reach the detector and therefore a 17 % chance that two emitted together will both reach the detector. The further the source is from the detector, the less likely it is that the two gamma-rays will be detected together (1.5 % solid angle and 0.02 % chance of summing for our distant source). Note that at any source-to-detector distance there will be some degree of summing (Figure 8.4). However, beyond a certain distance, which

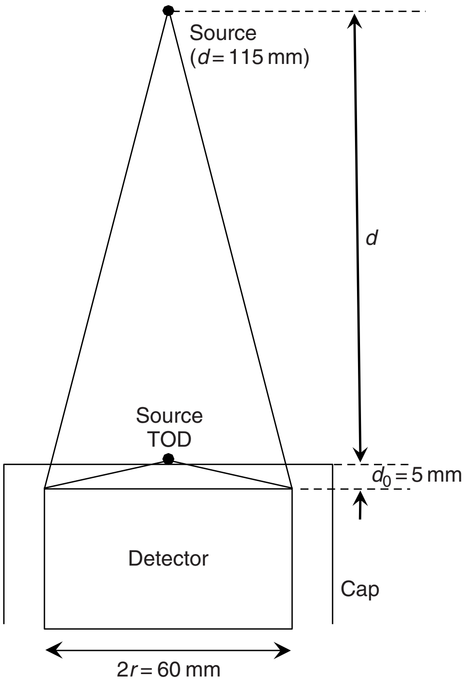

Figure 8.3 Geometrical arrangements used to obtain the data shown in Figure 8.1

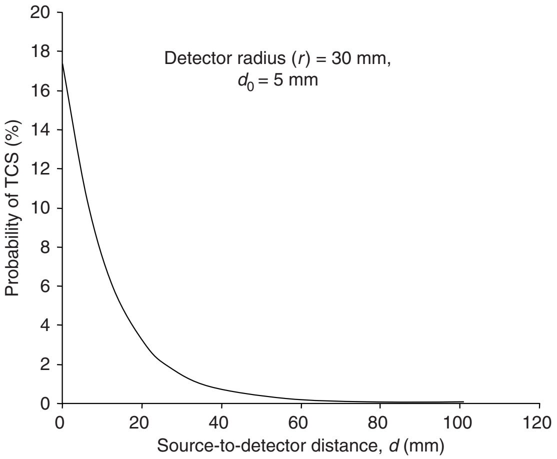

Figure 8.4 Calculated probability of summing as a function of source-to-detector distance: detector radius, $r = 30\,\mathrm{mm}$; $d_0 = 5\,\mathrm{mm}$

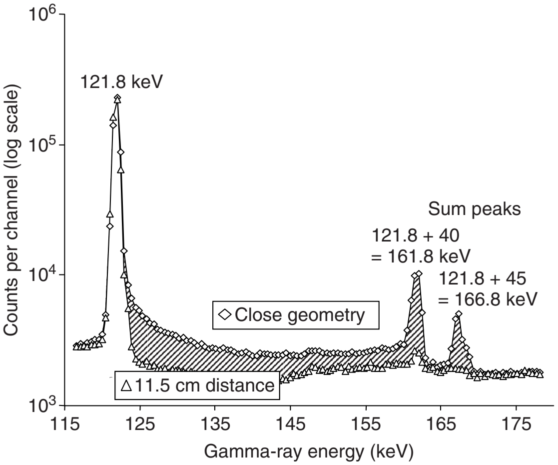

Figure 8.5 Sum peaks above the 121.78 keV peak of ^{152}Eu. Note also the raised level of the continuum at the high-energy side of the peak

depends upon the detector size, TCS losses will be negligible in practice.

A rough and ready estimate of the likelihood of summing of two photons emitted at the same time, P, can be estimated by using the following equation:

$$P = (R - D)^2/2R^2 \tag{8.1}$$

where R is the radius of the sphere into which the source emits and $D = d + d_0$, with d being the measured source-to-detector distance and d_0 the distance between detector face and the detector cap. If r is the detector radius, then $R = \sqrt{(r^2 + D^2)}$. The probability of one photon passing through the detector is the ratio of the solid angle subtended at the detector, $2\pi R(R - D)$, to the total area of the sphere, $4\pi R^2$. The probability of two photons emitted at the same time striking the detector is that probability squared.

It is worth noting that, for a given solid angle, the number of true coincidence summing events per second (but not the ratio of lost/total counts) will be directly proportional to the sample activity. On the other hand, random summing losses are a function of the *square* of the sample activity. In the situation in Figure 8.1, we can be quite sure that the problem with the 'top-of-detector' count is due to TCS, rather than random summing, because the measurements had almost the same count rate.

8.4 SPECTRAL EVIDENCE OF SUMMING

Since every de-excitation of ^{152}Sm, whatever gamma-rays are emitted, is likely to produce an X-ray these play a prominent role in TCS. Figure 8.5 shows portions of the spectra from which the calibration curves of Figure 8.1 were calculated. Comparing the two spectra reveals the presence of pairs of small sum peaks approximately 39.9 and 45.3 keV higher in energy than the expected peaks. This is emphasized by the partial analysis of the low-energy portion of the close geometry spectrum reproduced in Table 8.1.

Table 8.1 Partial analysis of the close geometry spectrum of ^{152}Eu

Energy (keV)[a]	Area (cps)	RSD (%)	Attribution (energies in keV)
39.60	386.00	0.46	Sm Kα X-rays
45.23	109.60	1.00	Sm Kβ X-rays
121.83	469.20	0.30	—
161.62	19.40	3.20	Sum 121.78 + 39.91
167.16	6.00	8.64	Sum 121.78 + 45.4
244.83	63.80	0.98	—
284.70	2.60	15.01	Sum 244.70 + 39.91
290.17	1.40	26.54	Sum 244.70 + 45.4
296.09	3.00	10.34	—
344.44	225.10	0.41	—
367.33	10.20	4.85	Sum 121.78 + 244.70
411.28	13.20	2.70	—
444.12	14.40	2.52	—
488.79	1.80	13.51	—
564.29	2.60	13.32	Sum 121.78 + 443.97

[a] Energies here are as reported by the analysis program.

All the peaks not attributed in Table 8.1 are normal full-energy peaks of ^{152}Eu. The extra peaks due to summing with the X-rays are clearly identifiable, appearing after each major gamma-ray originating from the electron-capture decay. In addition, we can detect γ–γ coincidences between some of the ^{152}Sm gamma-rays with higher abundances. The independence of the two branches of the ^{152}Eu decay is underlined by the fact that there is no peak at 466.06 keV, which would indicate summing between the 121.78 and the 344.28 keV gamma-rays, because these originate in different cascades. Nor is there summing between the Sm branch X-rays and the Gd branch gamma-rays. However, elsewhere in the spectrum the $(344.28 + 778.90)$ sum peak does appear as a consequence of summing within the beta decay branch.

Each sum peak represents only some of the counts lost from the main peaks – only some because there will also be a chance of summing in the detector with each and every gamma-ray in the cascade whether or not fully absorbed. In fact, since only a minority of gamma-rays are fully absorbed, the summing of a gamma-ray destined for a full-energy peak with an incompletely absorbed gamma-ray is more unlikely. As we shall see, these coincidences with partially absorbed gamma-rays must be taken into account if a TCS correction is to be computed.

Figure 8.5 also demonstrates a feature of summing that only occurs in p-type detectors. In the summed spectrum, the level of the background continuum at the high-energy side of the peaks is higher than that at the low energy side and the peak has a pronounced tail. Quite the reverse from the peak background we would normally expect, which is lower on the high-energy side (see Chapter 9, Figure 9.6). This phenomenon has been identified by Arnold and Sima (2004) as consequence of the electron capture X-rays generating a partial signal, in coincidence with the gamma-rays, within the dead layer of the detector, which may not be as 'dead' as thought. They suggest that the thick outer n+ contact layer diffuses some way into the high-purity germanium, creating a transition zone. That zone is capable of detecting the X-rays but with poor charge collection. These partial signals are then able to sum with the gamma-rays, creating the tail on the high-energy side of each peak. The effect does not happen with n-type detectors, where the contact layer is very thin, nor is there tailing on gamma-rays from β^- decay because there are no coincident X-rays. For example, elsewhere in the spectrum from which Figure 8.5 was taken, the 778.9 keV gamma-ray of ^{152}Eu, which is emitted following the β^- decay branch to ^{152}Gd, is not tailed.

A further possibility that I perhaps ought to mention is that in the case of beta decay, because the beta particle and the de-excitation gamma-rays are emitted almost at the same instant, a gamma-ray may sum with the bremsstrahlung produced as the beta-particle is slowed down.

8.5 VALIDITY OF CLOSE GEOMETRY CALIBRATIONS

Returning to the efficiency calibration data in Figure 8.1, I can say, without equivocation, that all the points in the close geometry set are invalid as far as constructing a calibration curve is concerned. That is not to say that they are altogether useless. Each point represents a valid calibration point for ^{152}Eu measured on the particular detector, in the particular source geometry on the detector cap. Although valid for ^{152}Eu, the points have no relevance to any other nuclide. For example, the point representing the 121.8 keV efficiency could not be used to estimate the activity of a ^{57}Co source via the 122 keV peak area. ^{57}Co will have its own, different, TCS problems.

While the lower curve in Figure 8.1, measured at 115 mm, appears to be satisfactory, we cannot say that there is no summing. All we can say is that the degree of TCS is negligible. (In fact, Figure 8.5 shows that even in the 115 mm spectrum there is a small sum peak indicating a small degree of summing.) For the same source measured at the same distance on a larger detector, the summing would not necessarily be negligible.

8.5.1 Efficiency calibration using QCYK mixed nuclide sources

In the United Kingdom, the readily available multi-nuclide reference materials QCY and QCYK, supplied by Isotrak, a subsidiary of AEA Technology QSA, are often used for gamma spectrometer efficiency calibration. Similar materials are available elsewhere, possibly with slightly different mixtures of nuclides. The QCYK mixed nuclide reference material contains twelve nuclides emitting gamma-rays from 59.54 keV (^{241}Am) to 1836.05 keV (^{88}Y). However, it seems not to be appreciated that these sources contain a number of nuclides, ^{57}Co, ^{60}Co, ^{88}Y and ^{139}Ce, which will exhibit true coincidence summing if measured close to the detector, significantly affecting the efficiency calibration, as demonstrated in Figure 8.6. In this figure, the dashed line represents the true calibration line passing through the non-summed nuclides. Table 8.2 lists the nuclides in QCYK and comments on various spectral features that became evident when a particular source was measured close to an n-type HPGe detector. These features are:

- ^{60}Co and ^{88}Y both emit two gamma-rays and summing between their gamma-rays is to be expected. That will reduce the peak areas of the full energy peaks of those nuclides and give rise to a sum peak in the spectrum corresponding to the sum of the gamma-ray energies. In Figure 8.6, these points can clearly be seen to lie below the corrected calibration line.

- Seven of the twelve nuclides decay by electron capture and, as we saw in Section 1.2.3, every decay will be accompanied by X-rays characteristic of the daughter atom. That means that we can expect summing between the gamma-rays and the X-rays. As Table 8.2 indicates, γ–X sum peaks from ^{57}Co, ^{139}Ce and ^{88}Y are very evident (see Figures 8.7(b) and 8.7(c)). Although similar summing in ^{241}Am and ^{65}Zn can be detected, it was barely significant in the particular spectrum analysed.
- ^{57}Co is a special case in that although the 122.06 keV peak sums out due to summing with the electron-capture X-rays and with the 14.41 keV gamma-ray, the 136.47 peak sums *in*. This is not obvious in the spectrum but is noticeable in the efficiency calibration where the 122.06 keV peak lies below the calibration line and the 136.47 above.
- A number of minor peaks were observed that could be identified as sums of gamma-rays with the germanium escaped electron capture X-ray (column 6 in Table 8.2). Germanium escape peaks from the ^{241}Am gamma-ray were also detected (see Chapter 2, Section 2.2.1).
- Single and double escape peaks from the gamma-rays of greater than 1022 keV can be expected. Not all were observed, however.

We might expect γ–X summing from ^{109}Cd. After all, it is an electron capture nuclide and the low energy end of the spectrum is dominated by the intense X-ray peaks from the ^{109}Ag daughter. If that were so, there should be sum peaks at around 88.03 + 22 keV – there are none. The gamma-ray we normally attribute to ^{109}Cd is actually emitted by the 39.8 s half-life metastable state ^{109m}Ag. Because of this, there is a delay in emission of the gamma-ray and no true coincidence between the silver X-ray, emitted at the moment of decay, and the 88.03 keV gamma-ray. ^{109}Cd is, therefore, not subject to summing. Interestingly, however, in the particular spectrum measured, with a 4.5 % dead time, we did observe minor random summing peaks between the major higher-energy peaks and the 22 Ag K X-rays. For the same reason, ^{113}Sn is not subject to TCS even though its electron capture X-rays are clearly visible in the spectrum. In this case, the 391.70 keV gamma-ray is, in fact, emitted by the 1.66 h half-life metastable state ^{113m}In.

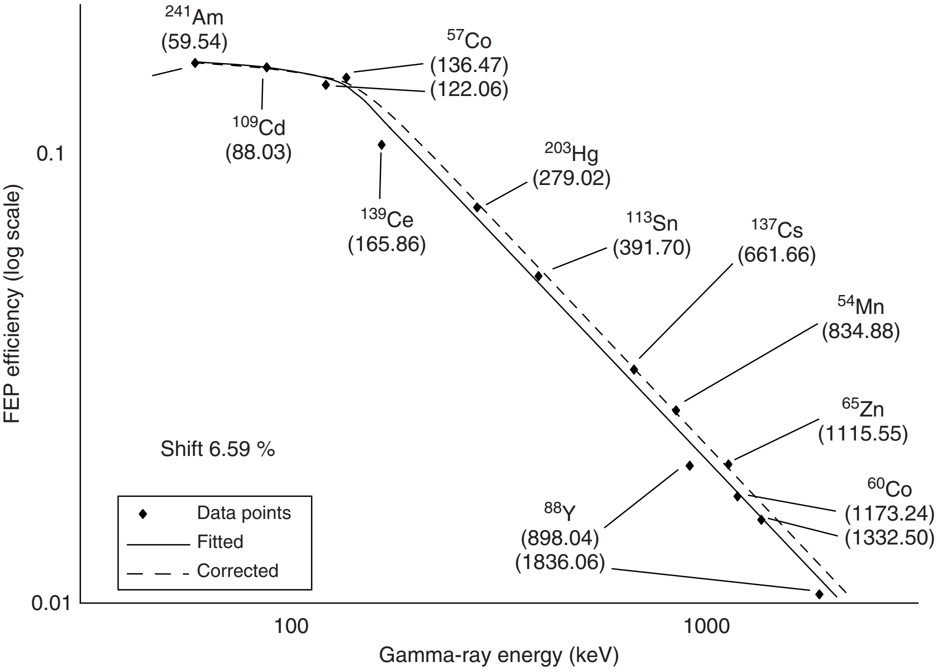

Figure 8.6 Close geometry efficiency calibration using the QCYK reference source

Table 8.2 Peaks observed within the QCYK spectrum measured at close geometry

Nuclide	Decay mode	X-rays	Gamma-ray energy (keV)	Summing[a]	Summing and Ge escape?	511 escape?[b]	Random-sum peak?[c]	Comments
^{241}Am	α	Np L	59.54	(γ–X)	—	—	—	Ge escape peaks
^{109}Cd	EC/IT	Ag K	88.03	None	—	—	—	No TCS – γ emission delayed by IT
^{57}Co	EC	Fe K	14.41	γ–γ	—	—	—	γ–X may be present but not resolved
			122.06	γ–γ and γ–X	✓	—	—	Sum with 14.41 with Ge escape
			136.47	γ–X	—	—	—	Sums in 122.06 + 14.41
^{139}Ce	EC	La K	165.86	γ–X	✓	—	—	—
^{203}Hg	β^-	Tl K (IC)	279.02	None	—	—	—	—
^{113}Sn	EC/IT	In X	391.70	None	—	—	—	No TCS – γ emission delayed by IT
^{85}Sr	EC	Rb K	514.00	γ–X	✓	—	—	Close to 511 – difficult deconvolution
^{137}Cs	β^-	Ba K (IC)	661.66	None	—	—	—	Possible SE from 1173.23 at 662.23
^{54}Mn	β^-	—	834.84	None	—	—	—	—
^{88}Y	EC	Sr K	898.04	γ–X and γ–γ	✓	—	—	—
			1836.05	γ–X and γ–γ	✓	S, D	✓	—
^{65}Zn	EC	Cu K	1115.54	(γ–X)	—	—	✓	—
^{60}Co	β^-	—	1173.23	}γ–γ	—	(S)	✓	—
			1332.49		—	D	✓	Close to SE from 1836.05 at 1325.05

[a] Parentheses indicates summing expected but not significant.

[b] S, single escape peak; D, double escape peak; Parentheses, indicate not observed.

[c] Random summing is with 22 keV Ag K X-rays from decay of ^{109}Cd.

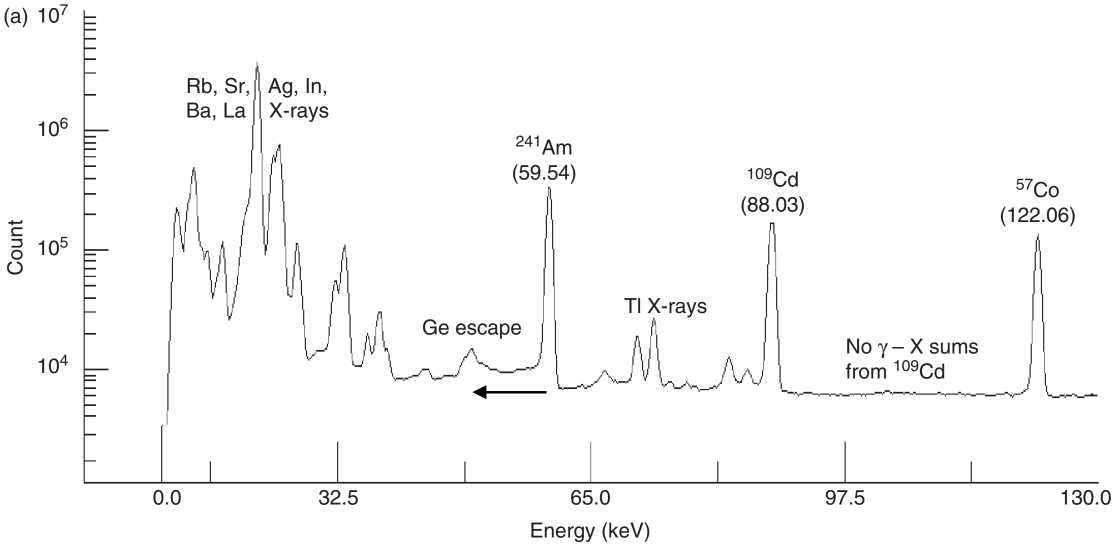

Figure 8.7 Parts of the QCYK spectrum at close geometry

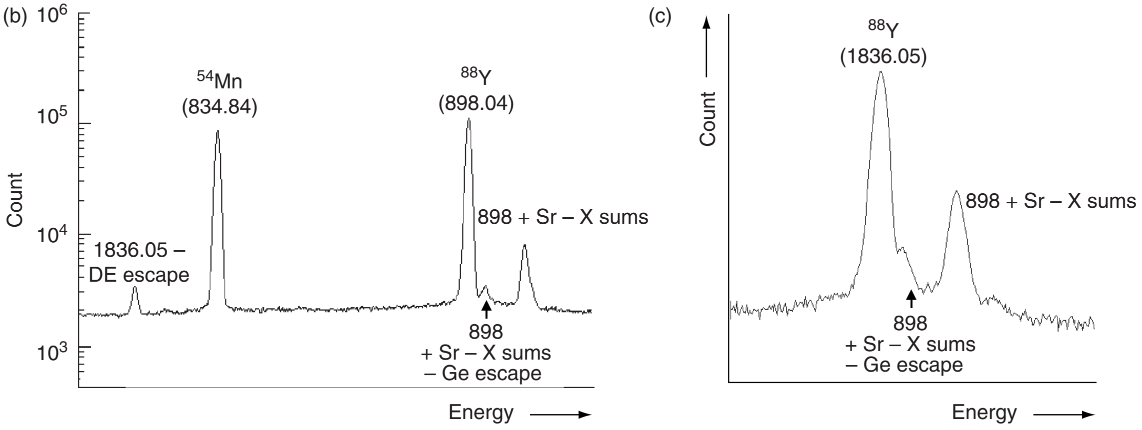

Figure 8.7 (Continued)

In the QCYK spectrum, there are a number of potential problems due to the close proximity of extraneous peaks to those to be measured. Particularly difficult is the measurement of the 514.00 keV peak of ^{85}Sr in the presence of the 511.00 keV annihilation peak. Unfortunately, most commercial software does not recognize the fact that the annihilation peak is Doppler broadened and deconvolution of the doublet may be questionable. A similar problem could arise when measuring the 1332.49 keV peak of ^{60}Co in the presence of the 1325.05 keV single escape peak of ^{88}Y, which will also be broadened. Fortunately, on any reasonable detector deconvolution will not be necessary. Attention can be drawn to the fact that the energy of the single escape peak of the ^{60}Co 1173.23 keV peak, 662.23 keV, is very close to the 661.66 keV peak of ^{137}Cs, although the intensity is very low and there is no problem in practice.

The effect of TCS in close geometry calibrations is to reduce the areas of the peaks due to ^{57}Co (the 136.47 keV peak is increased), ^{139}Ce, ^{88}Y and ^{60}Co. When the efficiency calibration is performed, the ‘best fit’ line will be somewhat lower than it should be (see Figure 8.6), meaning that activity estimates made using it will be a few percent in error. Nuclides that do not sum will be overestimated, while those that do sum will be underestimated by varying amounts. Whether that is acceptable or not depends upon the use to which the measured activities are to be put.

8.6 SUMMARY

I can summarize the essential points about TCS as follows:

- It usually results in lower full-energy peak areas (but see Section 8.8).
- It gets worse the closer the source is to the detector.
- It gets worse the larger the detector and is worst of all when using a well detector.
- It may be worse if a detector with a thin window is used because the X-rays that contribute to the summing will not be absorbed.
- It can be expected whenever nuclides with a complex decay scheme are measured.
- The degree of summing is not dependent upon count rate.

8.7 SUMMING IN ENVIRONMENTAL MEASUREMENTS

The last point in my summary is worth further discussion because of its importance in environmental measurements. Immediately after the Chernobyl accident, the gamma-spectrometric measurement of isotopes such as ^{137}Cs and ^{134}Cs was a major preoccupation for many laboratories. The sometimes heard opinion ‘*summing isn’t a problem for us – we only work at low count rates*’ is a dangerous simplification at best and probably utterly incorrect. ^{134}Cs has a relatively complex decay scheme and TCS is almost inevitable given that the environmental sample activities are low and must of necessity be measured close to the detector, often in Marinelli beakers. The extent to which TCS is ignored is best illustrated by Figure 8.8, which shows the results of an intercomparison arranged by the NPL in 1989 for the measurement of various radionuclides, including ^{134}Cs, at environmental levels. These broad features of the data were evident:

- Out of 58 ^{134}Cs results reported, only four were within the range expected by the NPL (i.e. within the shaded band in Figure 8.8.)

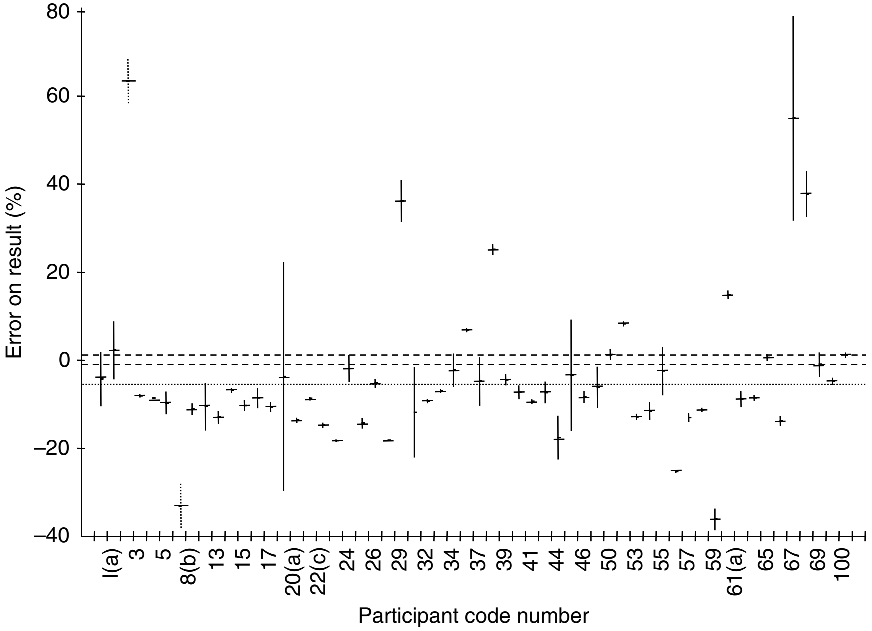

Figure 8.8 Summarized ^{134}Cs results from the 1989 NPL environmental radioactivity intercomparison exercise. (The band between the dashed lines represents the approximate 99 % confidence limit on the NPL source activity, the horizontal dashes mark the reported values and the vertical bars the reported 68 % confidence limits. Dashed vertical bars indicate that uncertainty was not reported)

- In only 11 cases did the true result lie within the 68% confidence limit reported by the measurement laboratory.
- It is obvious that the majority of the results were low; 64% of results reported were more than 5% below the expected value. (It is interesting to compare the magnitude of these errors with the calculated correction factors for ^{134}Cs given in Table 8.5 below.)

The most likely reason for this is that TCS had not been taken into account by the laboratories reporting these results. Bearing in mind that all the laboratories taking part in this intercomparison were reputable and believed that they were providing accurate results, Figure 8.8 was alarming. In fact, further NPL intercomparison exercises have been run regularly since then and, although some efforts do seem to be being made to correct for summing, the conclusion must be made that, in spite of the growing awareness of TCS, many laboratories still do not make appropriate corrections. Figure 8.9 shows similar data for measurements of ^{134}Cs taken from the 2002 NPL intercomparison. It is still the case that two thirds of the reported results are more than 5 % low while 50 % of the results can be judged as *significantly* low.

The message of this is obvious. True coincidence summing must be taken into account if accurate results are to be achieved. In defence of the gamma spectrometry community it should be said that until recently there were no easily usable tools for making TCS corrections and that in a busy laboratory it is not surprising that the time-consuming corrections were not made. A number of suggestions for making corrections and the modern software available are discussed below.

It is worth noting that, as important as it is in environmental measurement to achieve the lowest MDA, moving the sample a relatively small distance away from the detector will achieve a large decrease in TCS with only a small increase in MDA. The decrease in count rate due to larger source-to-detector distance is partly offset by fewer counts lost from peaks due to summing.

8.8 ACHIEVING VALID CLOSE GEOMETRY EFFICIENCY CALIBRATIONS

From what we have discussed it is evident that, leaving aside for the moment the possibility of making mathematical corrections, a valid calibration curve can only

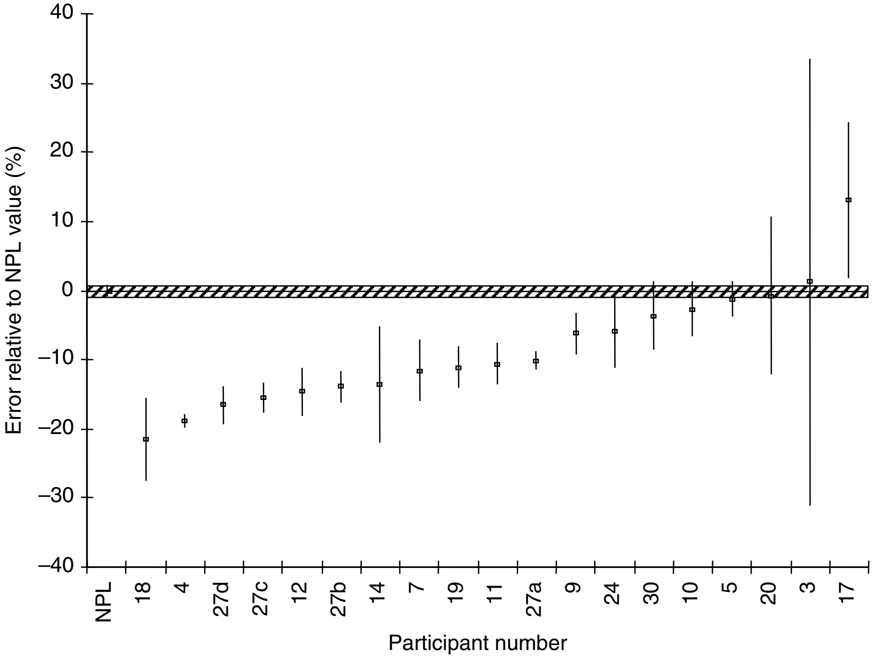

Figure 8.9 Summarized ^{134}Cs results from the 2002 NPL environmental radioactivity intercomparison exercise

be measured under close geometry conditions by using radionuclides which do not suffer from TCS. Table 8.3 suggests a number of nuclides that might be used. The list includes several that emit only a single gamma-ray and therefore can be expected not to participate in summing. However, care should still be taken with those single gamma-ray nuclides that decay by electron capture (e.g. ^{51}Cr). A detector with a thin window, and especially n-type detectors, may allow significant summing with the X-rays due to the lower absorption in the entrance window.

There are a number of nuclides which emit multiple gamma-rays but for which summing, of the specified gamma-ray, is usually negligible (e.g. ^{113}Sn and ^{131}I). Other gamma-rays are emitted with such a low abundance that summing, although possible, can be ignored. That might not be the case for a very large or a well detector.

Not all nuclides in the list are convenient to use in practice. The list includes some nuclides for which standardized sources may not be readily available. It also includes some sources with short half-lives that would not be appropriate for routine calibration. Nevertheless, with an appropriate selection of nuclides it is possible to create a valid close geometry efficiency calibration. There is, however, a scarcity of suitable nuclides providing gamma-rays above 1500 keV, thus preventing calibration at higher energy.

However, one must question what value such a calibration curve would have in practice. A large number of common nuclides have a complex decay scheme and are liable to exhibit the TCS problem. The carefully constructed TCS-free single gamma-ray efficiency curve will be irrelevant to the estimation of these nuclides. It will still be necessary to correct for the summing in the sample measurements. Overall, it is reasonable to suggest that the use of calibration curves for close geometry measurements is a waste of effort. The only value a summing-free close geometry calibration would have would be to help make the mathematical corrections described in Section 8.11.

In practice, we are usually constrained in the way that we handle our analysis by the software we use. It is pertinent to consider how the spectrum analysis program would treat calibration data such as that presented in Figure 8.1. Most likely it will blindly construct a 'best' (but completely invalid) fit, perhaps as shown in Figure 8.10. We know that all the points are likely to be lower than they should be and that the true calibration curve should lie above them all. (This is not universally true. For example, in the case of ^{134}Cs, summing of the 569.33 keV (15.38 %) and 795.83 keV (85.5 %) gamma-rays increases the area of the 1365.19 keV peak. This is called a **crossover transition**. The phenomenon is

Table 8.3 Radionuclides suitable for close geometry efficiency calibrations

Nuclide	Gamma-ray energy (keV)	Nuclide type[a]	Standard available[b]	Data in Appendix B
^{7}Be	477.60	S	Y	—
^{40}K	1460.82	S	(Y)	—
^{42}K	1524.67	M	Y	—
^{51}Cr	320.08	SX	Y	Y
^{54}Mn	834.84	SX	Y	Y
^{57}Co	122.06, 136.47	MX	Y	Y
^{64}Cu	1345.77	SX	Y	—
^{65}Zn	1115.54	S(X)	Y	Y
^{95}Zr	724.19, 756.73	M	—	—
^{95}Nb	765.80	S	Y	Y
^{103}Ru	497.08	S	—	—
^{109}Cd(^{109m}Ag)	88.03	S	Y	Y
^{113}Sn (^{113m}In)	391.70	M	Y	Y
^{131}I	364.49, 636.99	M	Y	—
^{137}Cs	661.66	S	Y	Y
^{139}Ce	165.86	SX	Y	Y
^{141}Ce	145.44	S	Y	—
^{144}Ce	133.52	M	Y	—
^{198}Au	411.80	M	Y	Y
^{203}Hg	279.20	S	Y	Y
^{210}Pb	46.54	S	—	—
^{241}Am	59.54	M	Y	Y

[a] S indicates a nuclide emitting a single gamma-ray; M indicates a nuclide for which the gamma-ray mentioned is the major one and has little coincidence summing; X indicates that summing with the accompanying X-rays (or other low energy gamma-rays may be a problem on thin-window or n-type detectors.

[b] Standards for these nuclides are available from radionuclide standard suppliers.

referred to as **summing in**. In such a case, the true calibration curve could lie below the data point.)

8.9 TCS, GEOMETRY AND COMPOSITION

In Chapter 6, I discussed the effect of sample geometry on count rate. Once again, it would be useful to consider a practical example. Table 8.4 lists the peak areas measured when the same amount of ^{152}Eu was counted as a point source and when distributed in water and in sand. The distributed sources were 13 mm in diameter and 20 mm high and measured on the cap of a 45 % p-type HPGe detector. As one would expect, there is an obvious overall loss of count rate due to the lower effective solid angle of the distributed sources and a more pronounced loss of count rate in the low energy peaks.

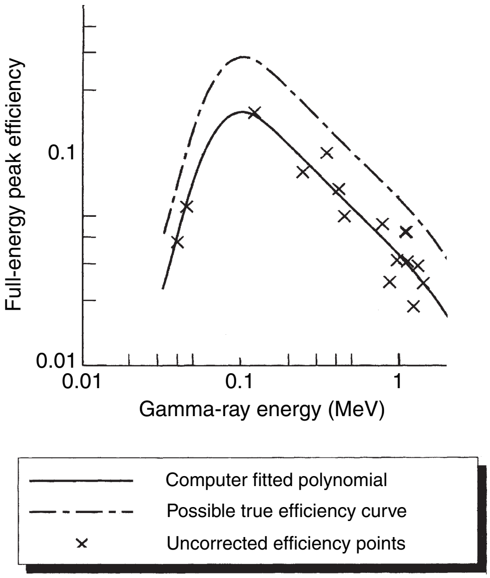

Figure 8.10 The efficiency curve constructed using a 'best fit' program

Table 8.4 Count rates of sources of different geometry[a]

	Peak area (cps)		
Energy (keV)	Point	Aqueous	Sand
39.91[b]	386.0	243.1	174.5
45.75[b]	109.6	75.0	58.1
121.78	469.3	325.6	301.0
244.70	63.7	46.4	44.8
344.28	255.1	151.8	139.0
411.12	13.2	9.51	8.62
443.97	14.4	11.0	10.6
778.90	46.6	32.7	30.2
867.38	9.67	7.26	7.23
964.07	35.6	26.4	25.5
1085.84	24.8	16.9	16.3
1112.08	30.4	22.8	22.3
1408.01	38.0	27.4	27.2

[a] All sources have the same amount (to within 0.5 %) of ^{152}Eu. Aqueous and sand sources are 13 mm in diameter and 20 mm high. All were measured in contact with the detector end-cap.

[b] Apparent energy of unresolved X-ray doublet.

Plotting the data graphically reveals more. Figure 8.11 shows the peaks areas for the distributed sources relative to the point source. The scatter of points, which cannot be attributed to peak area uncertainty, is at first unexpected but it is quite reproducible and is reminiscent of the scatter on the close geometry calibration. The explanation is straightforward. The ^{152}Eu in the point source is,

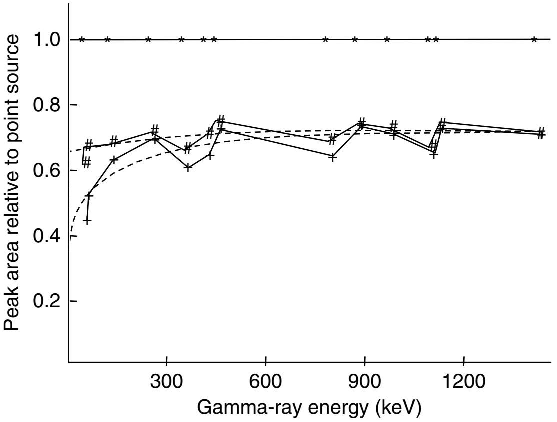

Figure 8.11 Relative peak areas for sources of different geometry and density: (∗) point source; (#) aqueous source; (+) sand source

on average, closer to the detector than in the distributed sources. The scatter represents the difference in TCS between the two source geometries. Moreover, the difference between the aqueous and sand matrix sources can, in addition to self-absorption, be attributed to the lower TCS in the sand source because of absorption of the Sm X-rays by the sand.

We can look further for evidence of the all-pervasive effects of TCS. Figure 6.9 (Chapter 6) relates the count rate of a source to its distance from the detector. The graphs confirm that the inverse square law can be applied to gamma spectrometry, at least in well-defined situations. A second look at the graphs reveals that the relationship becomes non-linear as the source nears the detector. The reason, of course, is the enhanced summing close to the detector.

The inescapable conclusion is that, unless sample and standard (or calibration) sources have identical shape and density, are in identical containers and are measured at the same distance, there will be differences in summing which will not be accounted for by the routine calibration process.

8.10 ACHIEVING ‘SUMMING-FREE’ MEASUREMENTS

It would seem that, if we must measure our samples close to the detector, our results would be in error due to TCS. It is important to remember that the degree of summing will be different for each nuclide and, if we are measuring more than one gamma-ray from a nuclide, different for each gamma-ray. Is there any way in which we can correct our results without resorting to the mathematical resolution described below in Section 8.11?

8.10.1 Using the ‘interpolative fit’ to correct for TCS

Both of the major gamma spectrum analysis programs allow a calibration option referred to as ‘Interpolative Fit’. For gamma-rays not in the calibration set, this option interpolates between the adjacent calibration points to estimate an efficiency value. For most gamma-rays one is likely to measure, the value returned will be in error. However, for those nuclides in the calibration set, exactly the correct value, which takes into account summing, will be generated. There is scope there, using the interpolative function, for creating efficiency curves using all of the nuclides one is ever likely to measure which will have an automatic correction for summing built in – at the expense of a great deal of effort and a large number of certified reference sources.

As an example, take the measurement of the NORM nuclides (Chapter 16). Most of the nuclides to be measured, the ^{238}U and ^{232}Th decay series, have complicated decay schemes and suffer from TCS, seriously in some cases. If reference materials containing the relevant nuclides (IAEA RGU-1 and RGTh-1, come to mind) were to be used as calibration standards, then effective efficiency data could be acquired. The efficiency curve would not be a pretty sight, because of the TCS, but as long as the interpolative mode was used, the correct, TCS-accounted-for, efficiency data would be used when analysing the sample spectra. It would not be acceptable, though, to use that calibration for measurements of any nuclides other than those represented in the calibration data.

8.10.2 Comparative activity measurements

The intercomparison data in Figure 8.8 reveals a small number of points that lie within the uncertainty expected by the NPL. There is nothing remarkable about that. In at least two of these cases (one of which I have particular knowledge!), the reason for the close agreement is that the measurements were made *comparatively*. The sample was compared directly with a calibrated source of ^{134}Cs measured in the same geometry at the same distance from the detector. In that way, the TCS errors are the same for sample and standard and cancel out. There is no need for a calibration curve at all. This would seem to be the most direct way of avoiding calibration errors due to TCS and, unless there are specific reasons to do otherwise, I would recommend direct comparison with standards for measurements at close geometry. (This is, of course, in effect, what the interpolative fit referred to above is achieving.)

8.10.3 Using correction factors derived from efficiency calibration curves

Comparative analysis does pose logistic problems. Not least is the fact that the spectrum analysis program may insist that results are calculated by reference to a calibration curve. The only option then is to derive, by measuring reference sources, a set of correction factors for each gamma-ray to be measured that can be applied to the output from the program. (Indeed, in this situation, since the initial results are known to be wrong because of the inadequacy of the calibration curve, there is little point in measuring one at all. Any calibration data at all could be used as long as the correction factors were consistent with it.) Taking as an example the data in Figure 8.10, the procedure would be as follows:

- Construct a notional efficiency calibration.
- Measure calibrated sources of each of the nuclides to be determined, prepared in the standard geometry and of the same density as the samples to be measured.
- Use the computer program to calculate the activity of these sources based upon the notional calibration. The ratio between measured and actual activities is the correction factor to be applied to the sample measurements.

It is important to note that, if the final result is to be based upon more than one gamma-ray, it will be necessary to make a correction to the result from each individual gamma-ray before combining to achieve the final corrected result. If standardized reference sources are not available for a particular nuclide that must be measured, a direct measurement of the correction factor is not possible. In that case, it would be necessary to perform measurements at a large source-to-detector distance using an appropriate TCS-free calibration curve and compare these with the close geometry measurements.

8.10.4 Correction of results using ‘bodged’ nuclear data[1]

Having measured the correction factors as described above, it is then necessary to correct gamma-ray intensities for TCS, combining results for different gamma-rays of the same nuclides, in order to achieve a satisfactory measurement. Although it is any easy task to set up in a spreadsheet, it is inconvenient for routine operation of a spectrometer system. It is much more acceptable if the system can be ‘tweaked’ so that the results from the program are correct. This can be achieved in a roundabout way.

Calculation of activity involves dividing the peak count rate by the gamma emission probability and by the efficiency. If the gamma emission probability in the nuclide library is adjusted by multiplying it by the TCS correction factor (i.e. the factor that the measured result would be multiplied by to achieve the correct result) then, because the efficiency is incorrect by that same amount, but in the opposite sense, then the calculated result must be correct. Note that this adjusted library must only be used for samples, not for the calibrations. There would have to be a separate adjusted library for each detector and each sample geometry. There is obvious scope for confusion. Nevertheless, this does appear to be the simplest means of coping with TCS corrections on a routine basis if activities must be calculated by reference to an efficiency curve.

8.11 MATHEMATICAL SUMMING CORRECTIONS

In principle, it is possible to correct for TCS errors mathematically. Take the simplest possible decay scheme in which we could expect TCS in Figure 8.12. The beta decay to one of two excited states is followed by the emission of the three gamma-rays shown. To simplify matters for the purposes of illustration, assume that the internal conversion coefficients for the gamma-rays are all zero. If the source activity were A Becquerels, in the absence of TCS, the count rate in the full-energy peak 1 would be:

$$n_1 = Ap_1\varepsilon_1 \tag{8.2}$$

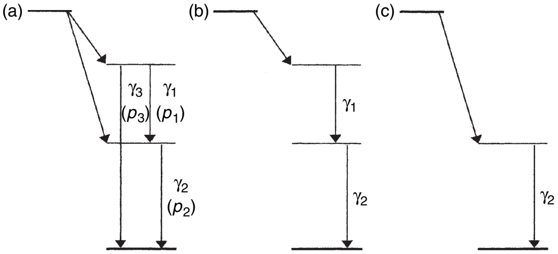

Figure 8.12 Simple illustrative decay schemes liable to true coincidence summing

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[1] ‘Bodge’ is British slang for a clumsy, messy, inelegant or inadequate solution to a problem.

with p_1 and ε_1 being the gamma emission probability and the full-energy peak efficiency of detection of γ_1, respectively. Similar equations, with the appropriate p and ε, would be used to calculate the full-energy peak count rates for γ_2 and γ_3.

Now, we will lose counts from the γ_1 peak by summing with γ_2. We need not consider γ_3 as de-excitation of the upper level can only give γ_1 or γ_3 – not both – and we only need the partial decay scheme in Figure 8.12(b). The number of counts lost (per second) by summing can be calculated as the product of:

the number of atoms decaying (A)

× the probability of de-excitation producing γ_1 (p_1)
× the probability of γ_1 being detected and appearing in the full-energy peak (ε_1)
× the probability of γ_2 being detected and appearing anywhere in the spectrum (ε_{T2})

I noted elsewhere that we must account for all coincidences whether giving rise to a sum peak count or not and hence, the final term uses ε_{T2}, the total efficiency for the detection of γ_2. Therefore, the net peak area would be:

$$n_1' = Ap_1\varepsilon_1 - Ap_1\varepsilon_1\varepsilon_{T2} \tag{8.3}$$

The ratio n_1/n_1' would then be used to correct for the TCS losses of the γ_1 peak area.

For γ_2, the situation is slightly different in that not all gamma-rays emanating from the intermediate energy level are a consequence of the de-excitation from the higher level. Some are preceded immediately by the β^- decay and cannot contribute to summing (see Figure 8.11(c)). The number of summing events is the product of:

the number of events giving rise to γ_2 (Ap_1)

× the probability of detection of γ_2 in the full-energy peak (ε_2)
× the probability of the detection of γ_1 anywhere in the spectrum (ε_{T1}).

and so the net area of peak 2 would be:

$$n_2' = Ap_2\varepsilon_2 - Ap_1\varepsilon_2\varepsilon_{T2} \tag{8.4}$$

Every true summing event of completely absorbed gamma-rays will produce a count in a peak equivalent to the sum of the energies and so the peak corresponding to the crossover transition, γ_3, will be increased in area rather than decreased. Following the same reasoning as above, the net count rate would be:

$$n_3' = Ap_3\varepsilon_3 + Ap_1\varepsilon_2\varepsilon_3 \tag{8.5}$$

Often, it is the case that the crossover transition probability is small and, because the emission probabilities for the normal cascade transitions are high, the summing-in can be much greater that the direct emission. Unless taken into account, the error from using peak areas due to this transition would be large.

It must be emphasized that this is all a gross simplification. In general, we cannot depend upon the internal conversion coefficients being negligible and we have considered a much simpler decay scheme than we can normally expect. If we examine instead the ^{152}Eu decay scheme (which is more representative of the real situation), the enormity of the task of correction for TCS becomes apparent. Not only must we take into account every possible coincidence, every cascade on the ^{152}Sm side of the scheme is likely to be in coincidence with the Sm X-rays emitted.

There are further complications if the source emits positrons. The 511 keV annihilation quanta can appear in coincidence with the gamma-rays from de-excitation of the daughter nucleus. An analytical solution is to add a pseudo energy level to the decay scheme 511 keV above the level in which the positron emission leaves the daughter. It is then necessary to assign a conversion coefficient of -0.5 to the pseudo level to take into account both of the annihilation photons. We could also complicate matters even further by taking into account triple coincidences between the most intense gamma-ray and bremsstrahlung coincidences.

The mathematics become more complicated if we consider *real samples* that might have considerable size and may have a composition such that significant self-absorption within the sample occurs. We know that electron capture X-rays play a significant role in the summing process. Self-absorption will mean that those X-rays, and low energy gamma-rays, will be absorbed more than higher energy gamma-rays altering the correction factors. For accurate coincidence correction, either we need separate efficiency information for each sample geometry and composition or the mathematics must take into account self-absorption.

Of course, in order to perform these calculations at all we must have available a full-energy peak efficiency free of TCS errors *and* a total efficiency curve. The task is daunting; nevertheless, in principle, if we have available the detailed decay scheme, adequate full-energy peak efficiency data, adequate total efficiency data, complete

Table 8.5 Examples of calculated TCS correction factors for four detector/source arrangements (data abstracted from Debertin and Schötzig (1990))[a]

Energy (keV)	Correction factor (multiplier)			
	A2	B2	A4	C
^{152}*Eu*				
39.91	1.349	1.559	1.079	—
121.78	1.262	1.648	1.058	1.13
244.70	1.434	2.086	1.088	1.18
344.28	1.146	1.145	1.037	1.07
411.12	1.424	1.432	1.075	—
443.97	1.378	2.373	1.096	1.16
778.90	1.249	1.256	1.045	1.13
964.07	1.249	1.438	1.035	1.10
1085.84[b]	0.940	1.177	0.992	0.97
1112.08	1.182	1.709	1.035	1.07
1408.01	1.208	1.790	1.038	1.08
^{134}*Cs*				
604.72	1.252	1.249	1.063	1.13
795.83	1.265	1.258	1.055	1.13
1365.19[b]	0.839	0.761	0.975	0.85

[a] Detector – source geometry: A, 12.5 % Ge(Li); A2, point source on detector cap; A4, 1 L Marinelli beaker: B, 25 % n-type HPGe; B2, point source on detector cap: C, 30 % Ge(Li), 24 mm diameter, 24 mm high source.

[b] Crossover transitions.

conversion coefficient data and detailed knowledge of the shape and composition of the sample, then a mathematical correction is possible. Table 8.5 lists TCS correction factors for a number of major gamma-rays of ^{152}Eu and ^{134}Cs, calculated by Debertin and Schötzig (1990) using the principles described above. A number of points stand out:

- The magnitude of the factors is consistent with the scatter in the close geometry curve in Figure 8.1 and with many of the errors apparent in Figures 8.8 and 8.9.
- The larger ^{152}Eu factors for the n-type detector (B2) can be attributed to extra summing because of the lower absorption of the Sm X-rays in this type of detector.
- The enhancement of the crossover transitions (1085.84 keV in ^{152}Eu and 1365.19 keV in ^{134}Cs) is apparent by the fact that their correction factors are less than one.
- The larger factors for the point sources (A2, B2) compared to the distributed sources (A4, C) is as expected for sources that are effectively closer to the source.

The paper from which the data were taken and a similar paper by Sinkko and Abalone (1985) contain useful compilations of correction factors which provide a general guide as to the likely importance of TCS for a large number of common nuclides. I must emphasize, however, that these correction factors should not be used to correct one's own data. They relate only to the particular detector and particular source geometries for which they were calculated. Correction factors for one's own data can only be determined by actual measurements or by reproducing the calculations described in the papers with the appropriate data for the detector system used. Unfortunately, while there are a number of well-known computer programs available for calculating TCS corrections (see the Reading List at the end of this chapter), these are not easily used in conjunction with the commonly used spectrum analysis programs.

One of the difficulties faced when collecting the information needed to perform reliable summing corrections is the reliability of the nuclear data in the literature. Of course, the quality of the data is worst for those nuclides for which summing is a particular problem. I can only remind the reader once more of the data given in Appendix B.

In Chapter 6, I mentioned the moves being made in the direction of providing detector calibrations based upon the dimensions and physical parameters of the detector and its encapsulation. Once this happy state of affairs has been attained, then the extension of that to include true coincidence corrections becomes possible. Again, one can envisage a situation where a new detector is delivered with a CD-ROM holding the theoretical calibration curve and the latest nuclear data for the nuclides of most common interest that would be accessed by spectrum-analysis programs for automatic summing corrections. For that, we must wait, but help does now appear to be on the horizon.

8.12 SOFTWARE FOR CORRECTION OF TCS

While the discussion above demonstrated the principle of calculating TCS corrections, in practice the situation is complex. It must be borne in mind that the corrections will depend upon sample distance, sample shape and, because of self-absorption, sample composition. Many attempts at making such corrections use Monte Carlo methods of calculation that consider the fate that many thousands of gamma-rays emitted from different parts of the source, impinging on different parts of the detector, might suffer, taking into account absorption within the source and in the detector. The program might construct an effective efficiency curve or may simply calculate correction factors to be applied to results from gamma-rays measured using the routine efficiency calibration. Both approaches have their advantages.

Monte Carlo methods demand a detailed knowledge of the detector geometry and construction. This information is not always available, that from the manufacturer being a nominal or estimated value. It is common, therefore, to read that Monte Carlo methods need 'fine tuning' with modification of parameters such as dead layer thickness and even detector diameter to make the model fit experimentally determined data. In some cases, people have resorted to X-raying their detector within its cooled encapsulation in order to measure the true detector size under operating conditions.

8.12.1 GESPECOR

GESPECOR (Germanium Spectrometry Correction Software) is one such Monte Carlo-based program which is commercially available. It was first created in 1996–1997 and has been under continuous development since, taking into account more and more minutiae of the detection process, including the partial dead layer phenomenon in p-type detectors referred to in Section 8.4. The program has been applied to all type of detector, including well types, and to all types of sample geometry – cylindrical, Marinelli and even 220 L waste drums. The program is undoubtedly worthwhile but it still not integrated into the spectrum acquisition process, meaning that there has to be some post-analysis correction to the result from each gamma-ray. In discussions with people who use GESPECOR and similar programs, they have warned that even minor inaccuracies in such factors as sample dimensions and composition can generate invalid results. Bearing in mind that in a routine gamma spectrometry laboratory, where often the composition of the sample is not well known and a separate calculation for each and every sample might be needed, this sensitivity is a drawback.

Normally one would not normally use sum peaks for efficiency calibration purposes. However, Arnold and Sima (20004) have shown how GESPECOR can be used to correct the sum peak intensities of ^{88}Y and ^{60}Co to extend the efficiency calibration energy range up to 2.7 MeV.

8.12.2 Calibrations using summing nuclides

I emphasized earlier that valid efficiency calibrations at close geometry can only be made directly using non-summing nuclides. For conventional calibrations, that is true, but Menno Blaauw (1993) has shown that it is possible to use the spectra of nuclides that are affected by summing to generate calibrations and TCS corrections.

The principle is as follows. Consider a decay scheme with, say, three excited states. There will be six possible gamma-rays emitted. For each of these gamma-rays, there will be an equation for the detection probability similar to Equations (8.2) to (8.5) above, albeit taking proper account of internal conversion coefficient, etc. Most of those equations will involve one full-energy peak efficiency and one total efficiency. Sum peak equations will have two full-energy peak efficiencies. We have six equations but much more than six unknown efficiencies. However, if we take into account the fact that on a log–log scale, the peak-to-total efficiency ratio is almost linear, it is possible to reduce the information needed to calculate total efficiencies to just two parameters. Even better, it is usually the case in complicated decay schemes that some of the crossover transitions (equivalent to the sum peak energies) have negligible emission probabilities. This leads to further simplification of the equations, such that we may have fewer unknowns than equations. It is possible to end up in a situation where not only the true full-energy peak efficiencies can be deduced, but also the parameters of the peak-to-total relationship and the activities of the source nuclides. We may not even need calibrated sources!

The solution of the equations is not trivial, needing an iterative fitting process. The method has been applied by Blauuw using ^{82}Br with some success and has been extended to deal with Marinelli beaker geometry and well-detectors. A development of the method introduces a third efficiency curve, called the 'linear-to-square' curve, in addition to full-energy peak and peak-to-total curves, which accounts for the variation of efficiency over the source volume due to self-absorption and scattering in the sample. This version is now incorporated into ORTEC's GammaVision, where it is assumed that the efficiency calibration source will include ^{134}Cs. It is, however, still necessary to create separate calibrations for each sample geometry and, presumably, sample composition.

8.12.3 TCS correction in spectrum analysis programs

When the first edition of this book was written, there were no facilities built into any of the readily available commercial spectrum analysis programs to make mathematical corrections for TCS. That situation has changed but it is not yet clear whether any great use is being made of these facilities or indeed whether they produce satisfactory results. While both GammaVision and Genie 2000 include facilities for making these corrections, the manuals provide little assistance to the inexperienced user on how to configure the programs.

Van Sluijs *et al.* (2000) evaluated the performance of three spectrum analysis programs in use in different European activation analysis laboratories from the point of view of TCS correction. None of the programs were easily available commercial programs and the conclusion was that inconsistencies between the programs needed investigation. Arnold *et al.* (2004) used the IAEA 2002 intercomparison spectra (see Chapter 15, Section 15.5.3) to evaluate seven commercially available spectrum analysis programs. Of these, only GammaVision and Genie 2000 could handle TCS corrections. They also used the external correction program, GESPECOR, to calculate correction factors. Where comparison between the three programs was possible, the correction factors were reasonably consistent. However, the results for GammaVision and Genie 2000 were not greatly better than for the other programs, especially for the spectra of uranium and thorium in equilibrium with their daughters. It would appear that there is no program available that can be trusted with TCS correction at the present time. It should be remembered that, depending on nuclide and source-to-detector geometry, TCS errors might be little more than 5 %. If the correction program produces results with an extra 10 % uncertainty, we would be little better off.

Even with a suitable program available, the TCS correction procedure will need some sort of extra calibration spectrum in order to derive a total efficiency versus energy relationship. The Genie 2000 system needs a number of nuclide sources, each emitting a single gamma-ray to cover the calibration range. The GammaVision calibration demands a single source containing a number of such nuclides and in addition, as mentioned above, a nuclide such as ^{134}Cs which suffers severe coincidence summing. From that single spectrum, GammaVision manages to derive all of the total efficiency data it needs. While it is easy, if expensive, to acquire appropriate single nuclide sources, there are few providers who can supply a traceable calibrated mixed source for the GammaVision calibration 'off-the-shelf'. However, recent work by Vidmar *et al.* (2005; 2006) may offer a solution. They have reported procedures for calculating both total efficiencies and the LS-curve for cylindrical samples, obviating the need for a practical measurement.

At the time of writing, facilities for TCS correction are beginning to become available but, as the results in Figures 8.8 and 8.9 suggest, are not being used routinely. Now that the process has started, perhaps we can hope that better, more user-friendly, programs, with adequate documentation, will be offered in the near future.

PRACTICAL POINTS

True coincidence summing:

- results in lower peak areas in general and possibly greater peak areas for crossover transitions;
- gets worse the closer the source is to the detector;
- gets worse the larger the detector and is worst of all when using a well-detector;
- may be worse if a detector with a thin window is used because more of the X-rays which might contribute to the summing will penetrate to the detector active volume;
- can be expected whenever nuclides with a complex decay scheme are measured;
- is not dependent upon count rate.

Accurate close geometry efficiency calibrations can only be achieved if nuclides are used which emit only one gamma-ray or if summing is accounted for. Whenever possible, close geometry calibration curves should be avoided by using direct comparison with an appropriate reference standard. If calibration curves cannot be avoided, correction factors must be derived for each gamma-ray of each nuclide used and the final results adjusted accordingly. It is important that that samples, standards and calibration sources have the same geometry and composition, or appropriate corrections are made.

In the last few years, a considerable amount of development effort has been put into programs that will allow TCS corrections to be made. In the near future, that effort may bear fruit.

FURTHER READING

- For a general understanding of summing errors, see:

Debertin, K. and Helmer, R.G. (1988). *Gamma- and X-ray Spectrometry with Semiconductor Detectors*, North Holland, Amsterdam, The Netherlands.

McFarland, R.C. (1993). Coincidence-summing considerations in the calibration of extended-range germanium detectors for filter-paper counting, *Radioact. Radiochem.*, **4**, 4–7.

- For a more detailed examination:

Sinkko, K. and Aaltonen, H. (1985). *Calculation of the true coincidence summing correction for different sample geometries in gamma-ray spectrometry*, Report STUK-B-VALO 40, Finnish Centre for Radiation and Nuclear Safety, Helsinki, Finland.

Debertin, K. and Schotig, U. (1990). *Bedeutung von Summationskrrektionen bei der Gammastrahlen-Spektrometne mit Germaniumdetektoren*, Report F Ra-24, Physikalisch-Technische Bundesanstalt, Braunschweig, Germany.

Debertin, K. and Schotzig, U (1979). Coincidence summing corrections in Ge(Li)-spectrometry at low source-to-detector distances, *Nucl. Instr. Meth. Phys. Res.*, **158**, 471–477.

- Calibrations using summing nuclides:

Blaauw, M. (1993). The use of sources emitting coincident γ-rays for determination of absolute efficiency curves of highly efficient Ge detectors, *Nucl. Instr. Meth. Phys. Res., A*, **332**, 493–500.

Blaauw, M. (1998). Calibration of the well-type germanium gamma-ray detector employing two gamma-ray spectra, *Nucl. Instr. Meth. Phys. Res., A*, **419**, 146–153.

Gelsema, S.J. (2001). *Advanced γ-ray spectrometry dealing with coincidences and attenuation effects*, phD Thesis, Interfaculty Reactor Institute, Delft University of Technology, The Netherlands.

Blaauw, M. and Gelsema, S.J. (2003). Cascade summing in gamma-ray spectrometry in Marinelli-beaker geometries: the third efficiency curve, *Nucl. Instr. Meth. Phys. Res., A*, **505**, 311–315.

- Evaluation of spectrum analysis programs:

van Sluijs, R., Bossus, D., Blaauw, M., Kennedy, G., De Wispelaere, A., van Lierde, S. and De Corte, F. (2000). Evaluation of three software programs for calculating true-coincidence summing correction factors, *J. Radioanal. Nucl. Chem.*, **244**, 675–680.

Arnold, D., Blaauw, M., Fazinic, S. and Kolotov, V.P. (2005). The 2002 IAEA intercomparison of software for low-level γ-ray spectrometry, *Nucl. Instr. Meth. Phys. Res., A*, **536**, 196–210.

- Assessments of some of the NPL intercomparisons are reported in:

Jerome, S. (1990). *Environmental radioactivity measurement intercomparison exercise*, Report RSA (EXT) 5, National Physical Laboratory, Teddington, UK.

Woods, D.H., Arinc, A., Dean, J.C.J., Pearce, A.K., Collins, S.M., Harms, A.V. and Stroak, A.J. (2003). *Environmental Radioactivity Comparison Exercise 2002*, NPL Report CAIR 1, National Physical Laboratory, HMSO, London, UK.

Arinc, A., Woods, D.H., Jerome, S.M., Collins, S.M., Pearce, A.K., Gillogan, C.R.D., Chari, K.V., Baker, M., Petrie, N.E., Stroak, A.J., Phillips, H.C. and Harms, A.V. (2004). *Environmental Radioactivity Comparison Exercise 2003*, NPL Report DQL-RN 001, National Physical Laboratory, HMSO, London, UK.

- Applications involving GESPECOR:

Sima, O. and Arnold, D. (2000). Accurate computation of coincidence summing corrections in low level gamma-ray spectrometry, *Appl. Radiat. Isotopes*, **53**, 51–56.

Arnold, D. and Sima, O. (2004). Application of GESPECOR software for the calculation of coincidence summing effects in special cases, *Appl. Radiat. Isotopes*, **60**, 167–172.

Arnold, D. and Sima, O. (2004). Extension of the efficiency calibration of germanium detectors using the GESPECOR software, *Appl. Radiat. Isotopes*, **61**, 117–121.

Sima, O., Cazan, I.L., Dinescu, L. and Arnold, D. (2004). Efficiency calibration of high volume samples using the GESPECOR software, *Appl. Radiat. Isotopes*, **61**, 123–127.

Information about the availability if GESPECOR can be found on the Internet at *http://www.matec-online.de.*

- On manual calculation of calibration curves:

Vidmar, T. and Likar, A. (2005). Calculation of total efficiencies of extended samples for HPGe detectors, *Nucl. Instr. Meth. Phys. Res., A*, **555**, 215–254.

Vidmar, T. and Korun, M. (2006). Calculation of LS-curves for coincidence summing corrections in gamma-ray spectrometry, *Nucl. Instr. Meth. Phys. Res., A*, **556**, 543–546.

- Test spectra that will allow TCS corrections to be tested:

Arnold, D., Blauuw, M., Fazinic, S. and Kolotov, V.P. (2005). The 2002 IAEA test spectra for low-level g-ray spectrometry software, *Nucl. Instr. Meth. Phys. Res., A*, **536**, 189–196.