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A New Measurement of the 60 keV Emission from Am-241 Using Metallic Magnetic Calorimeters

G. B. Kim[1] · S. T. P. Boyd[2] · R. H. Cantor[3] · A. S. Voyles[4] · J. T. Morrell[4] · L. A. Bernstein[5] · S. Friedrich[1]

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Abstract
We report a new measurement of the 60 keV transition from ^{241}Am. It uses a metallic magnetic calorimeter gamma-ray detector calibrated in the region around 60 keV by four accurately known X-rays and gamma rays from the decay of ^{169}Yb. We determine an energy of 59,539.3 ± 0.3 (stat) ± 0.3 (syst) eV, which is 1.6 ± 0.4 eV lower than the current literature value of 59,540.9 ± 0.1 eV. We discuss the sources of this uncertainty and approaches to address them.

Keywords Am-241 · Gamma-ray spectroscopy · Nuclear data · Metallic magnetic calorimeter · Microcalorimeter

1 Introduction

The 60 keV transition in the decay of ^{241}Am produces one of the most widely used calibration lines for low-energy gamma-ray detectors. ^{241}Am is long-lived and widely available, the transition has a high branching ratio of 35.9%, and the gamma-ray energy has an uncertainty of only ±0.1 eV, significantly smaller than that of most other isotopes [1, 2]. The current literature value of 59,540.9(1) eV has been determined from its difference to the ^{161}Tb gamma ray at 48,915.62(14) eV [2, 3], which in turn is based on a measurement with a bent crystal spectrometer [4]. The ^{241}Am–^{161}Tb spectra were measured repeatedly with one Si(Li) and two high-purity Ge detectors, and the top portion of the peaks were fit to a Gaussian function over a

✉ G. B. Kim
kim90@llnl.gov

1 Lawrence Livermore National Laboratory, Livermore, CA 94550, USA

2 University of New Mexico, Albuquerque, NM 87131, USA

3 Star Cryoelectronics, Santa Fe, NM 87508, USA

4 University of California, Berkeley, Berkeley, CA 94620, USA

5 Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

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"channel range of little more than one FWHM." The differences between the ^{241}Am and the ^{161}Tb centroids varied by 1 eV for the three detectors, and their weighted average had an uncertainty of 0.1 eV. Note that only the statistical uncertainties were included in [3]. Systematic errors, e.g., due to the low-energy tail of the detector response, the shape of the Compton background, the choice of the fit range or the nonlinearity of the multi-channel analyzer, have not been taken into account. While this is usually justified, the importance of ^{241}Am to calibrate low-energy gamma spectra makes it desirable to measure its decay radiation independently.

Metallic magnetic calorimeter (MMC) gamma-ray detectors [5] provide an order of magnitude higher energy resolution than semiconductor gamma-ray detectors and can therefore increase the accuracy of centroid measurements accordingly. They also have a predictable response function that is mostly linear with energy with only a small and reproducible second-order correction [6, 7]. This makes them well-suited to re-measure the energy of the 60 keV transition in the decay of ^{241}Am accurately. Among the possible calibration sources, ^{169}Yb is the best choice because its absolute gamma energies have been measured with an accuracy of ≤ 0.1 eV with a double-flat Si crystal spectrometer whose lattice spacing has been referenced to the Cs reference scale for frequencies [2, 8]. The K-shell X-rays of the ^{169}Yb daughter ^{169}Tm have been measured with similar accuracy [9, 10]. Since ^{169}Yb is not commercially available, we have produced it in a $(d, 2n)$ reaction by irradiation of a monoisotopic ^{169}Tm target with 15 MeV deuterons. This paper discusses our initial measurements of the 60 keV decay in ^{241}Am with an MMC gamma detector calibrated by ^{169}Yb.

2 Experiment

2.1 Yb-169 Calibration Source

Among all isotopes used for gamma detector calibration, the gamma rays of ^{169}Yb are known with the highest accuracy in the energy range below 300 keV [2, 8]. Specifically, the decay of ^{169}Yb produces a gamma ray at 63,120.44(3) eV with a branching ratio of 43.62% that is just above the energy of the ^{241}Am emission of interest. In addition, the ^{169}Yb decay generates strong Tm K X-rays whose energies are just below the ^{241}Am line and have been measured with similarly high accuracy [9, 10]. ^{169}Yb is therefore the ideal isotope to calibrate an MMC detector for an accurate measurement of the ^{241}Am gamma ray at 60 keV. Since ^{169}Yb is not commercially available, we made a 0.25 inch diameter target out of a commercial 100-μm-thick thulium foil, which consists to 100% of the isotope ^{169}Tm, and irradiated it at the 88″ Cyclotron at Lawrence Berkeley National Laboratory with a 15 MeV deuteron beam to produce ^{169}Yb. Simulations show that 15 MeV deuterons lose an average of 2.7 MeV in Tm over 100 μm. The beam energy was then chosen to produce ^{169}Yb with a cross section of ~500 mbarn throughout the foil in the reaction ^{169}Tm $(d, 2n)^{169}$Yb while minimizing the amount of ^{168}Tm, whose decay is accompanied by several strong gamma rays (Fig. 1). In addition, 15 MeV deuterons produce the stable isotopes ^{168}Yb and ^{170}Yb and the radioactive ^{170}Tm, whose decay produces an additional albeit weak calibration line at 84.25474(8) keV. For a beam

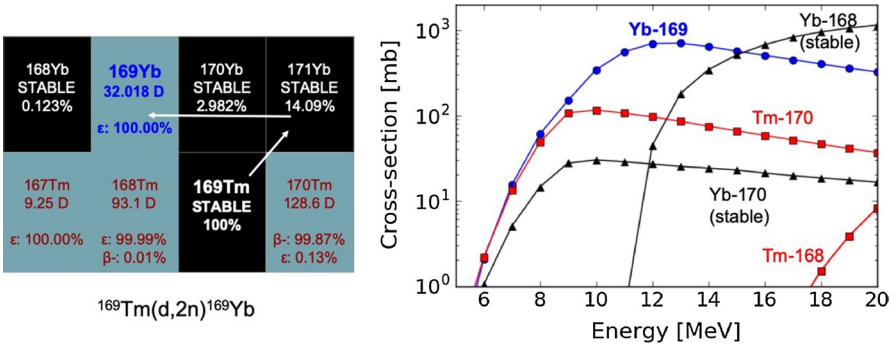

Fig. 1 (Left) Table of nuclides around ^{169}Yb [11]. The arrows indicate the way ^{169}Yb is produced in the (d, $2n$) nuclear reaction on ^{169}Tm. (Right) Calculated cross sections for a ^{169}Tm target as a function of deuteron energy [12]. ^{168}Tm is a radioactive isotope whose production should be minimized (Color figure online)

current of 750 nA, roughly a third of which hit the Tm target, we initially produced ~ 1.8 μCi of ^{169}Yb during one hour of irradiation.

2.2 Metallic Magnetic Calorimeters

The experiment used an MMC gamma detector array designed at the University of New Mexico (UNM) and fabricated at STAR Cryoelectronics [13, 14]. The MMCs consist of 30-μm-thick Au absorbers with an area of (500 μm)^2 that are supported on paramagnetic Ag/Er sensors by eight Au posts. They are operated in a cryogen-free dilution refrigerator with a base temperature < 7 mK. Gamma signals are amplified with a two-stage SQUID from STAR Cryoelectronics, with the first-stage SQUID being integrated on the same chip as the MMC. The array has 14 MMC pixels and has an energy resolution of as high as 38 eV FWHM at 60 keV [13]. The MMC response is very consistent with that of MMC gamma detectors developed at Heidelberg University and read out with a two-stage SQUID preamplifier from Magnicon [15]. This gives us some confidence that deviations from literature values are not due to peculiarities in the MMC response. For this experiment, we did not chemically separate the ^{169}Yb from the Tm target, but attached the irradiated target to a 1/16″ Cu foil to reduce low-energy Tm L X-rays and mounted it in front of the MMC gamma detector at a distance of 5 mm. An external ^{241}Am source is periodically added outside the cryostat for repeated measurements with and without ^{241}Am source to measure the background and check consistency.

2.3 Analysis Procedure

We capture the full gamma-induced waveforms with a four-channel 14-bit GaGe digitizer and write them to disk for subsequent off-line analysis. Only two pixels of the MMC array are selected for this measurement to avoid degrading the energy

resolution due to Joule heating of the MMC by power dissipation of the SQUIDs, which are located on the same substrate as the MMC [16]. We have found that a trapezoidal filter produces spectra with comparable energy resolution as an optimal filter that is typically used to process microcalorimeter signals [6] and prefer it because it is faster than the optimal filter and does not require template signals or noise spectra. Filtering parameters are set to a peaking time of 1 ms and a gap time of 10 µs. We correct for a drift of signal amplitudes due to slow temperature fluctuations by averaging 50 amplitudes of the 177.21307 keV line of ^{169}Yb and correcting all amplitudes by this scale factor.

3 Results

The combined gamma-ray spectra of the ^{169}Yb and ^{241}Am sources in the region around 60 keV are shown in Fig. 2. Since there is some line overlap of the ^{241}Am line of interest with the Tm K–$O_{2,3}$ X-ray at 59.3573(17) keV [17], we took spectra with and without the ^{241}Am source so that we could accurately determine the spectral background without the ^{241}Am line. ^{170}Yb X-rays from the ^{170}Tm decay are negligible in the spectrum. We determine the line shape from a strong gamma ray and apply it to other lines in the spectrum. We chose to extract the line shape from the isolated ^{169}Yb gamma ray at 177.2130 keV and fit it to a Gaussian function with small tails due to pile-up as before [7]. The small step due to small-angle scattering in the Cu filter is fit by a complementary error function centered at the peak position. We then keep the tail shape fixed and only vary the Gaussian parameters (amplitude, width, centroid), the step height and Gaussian-to-tail ratio, to fit the other gamma rays in the spectrum. For X-rays, we used a Voigt function to account for the natural X-ray linewidth.

Figure 3 shows the fits in the 60 keV region that includes the ^{241}Am gamma ray of interest and the two Tm $K_{\beta 2}$ and K–$O_{2,3}$ X-rays. They agree with the measured spectra within the statistical accuracy of the measurement. This suggests that there is no other significant Tm X-ray peak in the 60 keV region except the interfering Tm K–$O_{2,3}$ line.

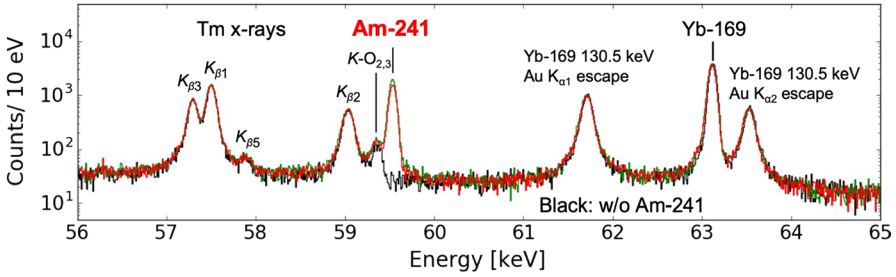

Fig. 2 Calibrated spectra from two MMC pixels. The measurement is repeated with (red, green for pixel 1, 2) and without (black) the ^{241}Am source (Color figure online)

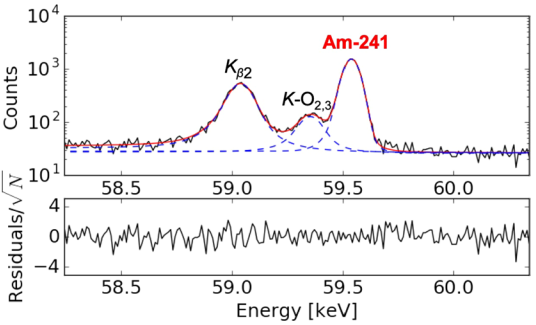

Fig. 3 Spectral fit to three peaks in the 60 keV region. Fits to individual peaks (dashed, blue) and the sum (solid, red) are shown (Color figure online)

The spectra are calibrated using only those X-rays and gamma rays that whose literature values are known with very high accuracy. This includes the Tm $K_{\beta 1}$ X-ray at 57.50876(15) keV [10], the ^{169}Yb gamma ray at 63.12044(3) keV [18], and the two Au K X-ray escape lines from the ^{169}Yb gamma ray at 130.52293(4) keV [18]. For a Au $K_{\alpha 1}$ energy of 68.80450(18) keV and a Au $K_{\alpha 2}$ energy of 66.99073(22) keV [10], these lines are seen at energies of 61.71843(18) and 63.5322(22) keV, respectively, with uncertainties and linewidths set by the Au K X-rays. Calibration uncertainties are obtained as before [6, 7] by successively varying the centroid energies by their statistical and literature uncertainties and calculating different calibration curves for each set of calibration points. The calibration uncertainty as a function of energy is then given by the standard deviation of these calibration curves (Fig. 4, shaded area).

Figure 4 shows the residuals of the energy calibration, i.e., the difference between the measured average energies from the two MMC pixels and their literature values. While the values of the calibration points are consistent with the literature values within the uncertainty of the measurement, the energy of the ^{241}Am gamma ray is

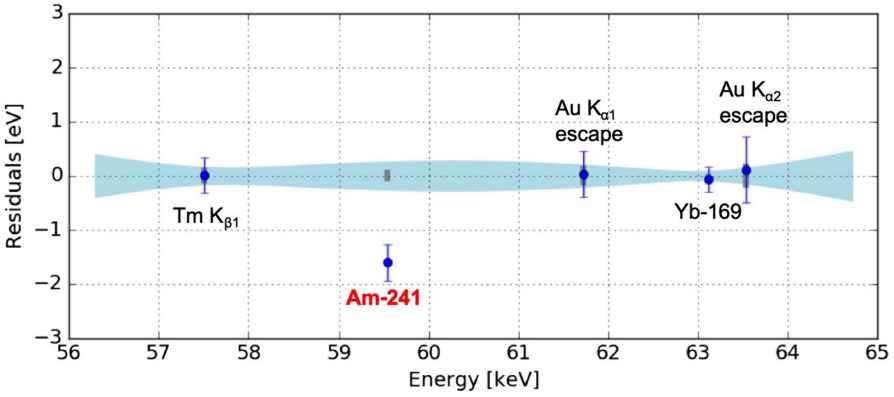

Fig. 4 Residuals of the energy calibration with statistical errors (blue bars), literature errors (gray bars) and total calibration errors (shaded area). While the residuals of the four calibration lines are consistent with zero, i.e., the measured energies are consistent with the literature values, the energy of the ^{241}Am emission is 1.6(4) eV smaller than the literature value of 59.5409(1) keV (Color figure online)

slightly lower. It is measured as 59.5393(4) keV, which is 1.6 eV lower than the current literature value of 59.5409(1) keV. This deviation is in the direction of an earlier evaluated value of 59.537(1) keV for the ^{241}Am emission [3]. Although the accuracy of the measurement is currently still limited by an error of 0.4 eV, the deviation is 4 σ, which suggests that the current literature value may be slightly off.

4 Discussion

MMC gamma-ray detectors are well-suited for accurate measurements of nuclear decay data due to their high energy resolution, their good linearity and their reproducible response function. We have combined MMCs with the most accurate low-energy gamma calibration source available, ^{169}Yb, to re-measure the energy of the important calibration line from the decay of ^{241}Am. We find an energy of 59.5393(4) keV that is 1.6(4) eV lower than the current literature value of 59.5409(1) keV. At this point, the 0.4 eV uncertainty of our measurement is still too high to firmly ascertain a discrepancy, and the experiment should therefore be repeated with more detectors and better statistics. In addition, there are systematic uncertainties of 0.3 eV in our measurement that have not been considered in the earlier characterization of the ^{241}Am decay. These include uncertainties due to detector drift, the choice of the fit function and range as well as binning effects. Finally, the precision of cryogenic detectors is reaching a point where the nonlinearity of ADC can make a non-negligible contribution to the observed nonlinearity. We are currently investigating ADC nonlinearities carefully [19], and may have to introduce an ADC correction to account for this effect. Once we understand all systematic errors quantitatively, it would ultimately be desirable to repeat this measurement at several institutions to assess if the systematic errors are confirmed by independent data sets. We are currently setting up a collaboration for that purpose.

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