256-pixel microcalorimeter array for high-resolution γ -ray spectroscopy of mixed-actinide materials

R. Winkler, A.S. Hoover, M.W. Rabin

Los Alamos National Laboratory, Los Alamos, NM

D.A. Bennett, W.B. Doriese, J.W. Fowler, J. Hays-Wehle, R.D. Horansky,
C.D. Reintsema, D.R. Schmidt, L.R. Vale, J.N. Ullom

National Institute of Standards and Technology, Boulder, CO

Abstract

The application of cryogenic microcalorimeter detectors to γ -ray spectroscopy allows for measurements with unprecedented energy resolution. These detectors are ideally suited for γ -ray spectroscopy applications for which the measurement quality is limited by the spectral overlap of many closely spaced transitions using conventional detector technologies. The non-destructive analysis of mixed-isotope Pu materials is one such application where the precision can be potentially improved utilizing microcalorimeter detectors compared to current state-of-the-art high-purity Ge detectors (HPGe). The LANL-NIST γ -ray spectrometer, a 256-pixel microcalorimeter array based on transition-edge sensors (TESs), was recently commissioned and used to collect data on a variety of Pu isotopic standards to characterize the instrument performance. These measurements represent the first time the simultaneous readout of all 256 pixels for measurements of mixed-isotope Pu materials has been achieved. The LANL-NIST γ -ray spectrometer has demonstrated an average pixel resolution of 55 eV full-width-at-half-maximum at 100 keV, nearly an order of magnitude better than HPGe detectors. Some challenges of the analysis of many-channel ultra-high resolution data and the techniques used to produce quality spectra for isotopic analysis will be presented. The LANL-NIST γ -ray spectrometer has also demonstrated stable operation and obtained high resolution measurements

Email address: rwinkler@lanl.gov (R. Winkler)
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at total array event rates beyond 1 kHz. For a total event rate of 1.25 kHz, approximately 5.6 cps/pixel, a 72.2 eV average FWHM for the 103 keV photopeak of ^{153}Gd was achieved.

Keywords: Microcalorimeter Gamma-ray High-Resolution Spectroscopy

Introduction

Detectors based on cryogenic microcalorimeter sensors have emerged as powerful tools for the detection of a variety of radiation types. These novel detectors are capable of photon and charged particle measurements with extraordinary energy resolution and have been essential for high-precision measurements in a multitude of spectroscopy applications. Experiments employing microcalorimeter detectors span a wide energy range; from sub-keV X -ray measurements to α -decay spectroscopy beyond an MeV [1–4]. In the realm of γ -ray spectroscopy, microcalorimeter detectors have achieved energy resolutions with FWHM resolution as low as 22 eV at 97.4 keV [5]. In the following, the operation and performance of the 256-pixel LANL-NIST microcalorimeter array and its application to difficult γ - and X -ray emission measurements are presented. While the applications for this instrument in γ -ray spectroscopy are plentiful, this article focuses on the high-precision, non-destructive analyses of complex, mixed-actinide materials. These material types emit a complex spectrum of both X - and γ -rays, particularly in the 100 keV region, that results in spectra containing many strongly overlapping peaks in measurements performed with state-of-the-art high-purity Germanium (HPGe) detectors.

In the realm of nuclear safeguards, a precise and accurate measurement of the intensity of γ -rays associated with the decay of various Pu isotopes is a vital ingredient in the determination of the relative Pu isotopic mass abundances. These relative mass abundances, such as the $^{240}\text{Pu}/^{239}\text{Pu}$ mass ratio, can be indicators of the purpose and manufacturing history of a mixed-isotope Pu material [6]. The photon emission spectra from Pu materials are complex, particularly in the 100 keV region, and the resolution of HPGe detectors can

57 limit the total measurement uncertainty of the determination of relative isotopic
 58 abundances within a mixed-actinide material. While the energy resolution of
 59 these semiconductor detectors is typically limited to a FWHM of approximately
 60 400 eV at 122 keV, the low-noise, low-temperature operation of arrays of mi-
 61 crocalorimeter detectors offers up to an order of magnitude improvement in
 62 energy resolution which in turn dramatically reduces spectral peak overlaps,
 63 allowing for high-precision measurements of complex X - and γ -ray emission.

Figure 1: (Color online) Photograph of the LANL-NIST γ -ray spectrometer. The 256-pixel array is comprised of four 64-pixel chips with a high-purity Sn absorber thermally attached to each TES. Each Sn absorber measures 1.5 mm on a side and has a thickness of 0.38 mm.

64 The microcalorimeter detectors of the LANL-NIST γ -ray spectrometer are
 65 based on superconducting transition-edge sensors (TESs) [7] consisting of a thin-
 66 film Cu-Mo bilayer with a normal-to-superconducting transition near 100 mK.
 67 When operated in the vicinity of this transition temperature, the resistance of
 68 the sensor is strongly dependent on its temperature. The thermalized energy
 69 of incident radiation absorbed by the detector increases the temperature of the
 70 sensor causing a substantial increase in its resistance. The TES is operated in a
 71 voltage biased mode [8] such that an increase in the resistance of the TES will
 72 lead to a decrease in the current flowing through the TES circuit that is propor-

73 tional to the energy of the thermalized radiation. The change in current flowing
 74 though a TES sensor is measured via an inductively-coupled Superconducting
 75 Quantum Interference Device (SQUID) amplifier. To improve the detection ef-
 76 ficiency of incident photons, a bulk Sn absorber is thermally coupled to the
 77 TES membrane. The thin Sn absorber yields a photopeak efficiency of 33% at
 78 100 keV compared to a value of nearly 100% for planar HPGe crystals where
 79 the greater stopping power is due to the increased detector thickness (typically
 80 between 1 and 1.5 cm).

81 **The 256-pixel LANL-NIST γ -ray spectrometer**

82 The 256-pixel LANL-NIST γ -ray spectrometer was recently fabricated and
 83 commissioned [9] for high-resolution γ -ray spectroscopy applications. The total
 84 active collection area of the array, shown in Figure 1, is 576 mm². This collection
 85 area is comparable to that of typical planar HPGe crystals of 200 to 1000 mm².
 86 A bulk 1.5 x 1.5 x 0.38 mm high-purity Sn absorber is thermally attached
 87 to each TES. Readout of pulse information is performed using a time-domain
 88 multiplexing scheme [10, 11] to minimize the number of physical connections to
 89 the instrument. In addition to decreasing the number of readout channels for the
 90 instrument, this also relaxes the difficult and expensive cryogenic requirements
 91 for operation at temperatures near 100 mK. Each TES of the array is coupled
 92 to a first stage SQUID in an 8-column, 32-row format. This first stage SQUID
 93 is operated in a flux-lock loop (FLL) to maintain linearity between the SQUID
 94 feedback signal and a change in the measured magnetic flux subsequent to the
 95 thermalization of an incident photon. Within each column, a row-address signal
 96 cycles through the 32 rows, sequentially applying bias to the first stage SQUID
 97 coupled to each TES. The case in which the signal provided by the digital
 98 feedback circuitry is insufficient to maintain operation of the SQUID amplifier
 99 within a linear response region is a so-called SQUID unlock event. In this case,
 100 a SQUID amplifier relock procedure is performed to re-establish linear amplifier
 101 response. The signals of a single row within each of the eight columns are read

102 out simultaneously. For each column, this signal is input into a single, shared
103 second stage SQUID amplifier. The output from the second stage SQUID is
104 then amplified by a SQUID series array [12] stage and finally by conventional
105 preamplifiers operated at room temperature.

106 The excellent energy resolution of a microcalorimeter detector is made pos-
107 sible by the careful selection of the combined heat capacity of the sensor and
108 bulk Sn absorber. The energy resolution and dynamic range of these detectors
109 are both functions of this combined heat capacity. The energy resolution is pro-
110 portional to the square root of the heat capacity of the detector while a decrease
111 in heat capacity decreases the dynamic range. Therefore there is a balance be-
112 tween desired energy resolution and the required dynamic range of a TES when
113 choosing the appropriate physical parameters of the sensor. This inhibits the
114 fabrication of arbitrarily large detectors and, for the high energy resolution de-
115 tection of 30 to 200 keV photons, limits the heat capacity of individual detectors
116 to approximately 22 pJ/K [9]. The array operating temperature of 85 mK is
117 maintained by an adiabatic demagnetization refrigerator (ADR) backed by a He
118 pulse tube cooler operating at approximately 3.5 K. These cryogenics provide
119 the means for the convenient and stable array operation at sub-1 K tempera-
120 tures without the use of liquid cryogenes. For a comprehensive description of
121 the fabrication, characterization, and commissioning of the LANL-NIST γ -ray
122 spectrometer, please refer to Bennett *et al.* [9].

123 The LANL-NIST γ -ray spectrometer was developed to improve the total
124 photopeak detection efficiency for photons below 210 keV compared to previous
125 array generations. For nuclear safeguards and material accountability appli-
126 cations, high-statistics data are crucial for the effectiveness of non-destructive
127 analysis techniques. The maximum achievable count rates of HPGe detectors
128 are typically up to 40 kHz before the energy resolution of measurements is sig-
129 nificantly degraded. The intrinsically slower recovery time of thermal detectors
130 compared to semiconductor types and the mode of multiplexing used for the
131 readout of the microcalorimeter array have traditionally limited the maximum
132 total event rate to below 1 kHz. Recent software and hardware upgrades of the

LANL-NIST γ -ray spectrometer readout electronics have been implemented to achieve the high-resolution operation of the array at total event rates beyond 1 kHz. In the following, the characterization of the high-rate operation of the 256-pixel microcalorimeter array and the effort to push for stable operation for event rates beyond 1 kHz will be described.

Characterization of the LANL-NIST γ -ray spectrometer

During recent commissioning measurements, the LANL-NIST γ -ray spectrometer demonstrated an average FWHM of 65 eV for the 97 keV photopeak emitted by a ^{153}Gd check source [9]. Similar fully multiplexed 8x32 measurements performed at Los Alamos National Laboratory have yielded the results depicted in Figure 2. At 97 keV, the average FWHM of the detectors of the spectrometer was determined to be 55 eV for individual pixel count rates of approximately 1 cps/pixel. For these measurements, spectroscopic data were obtained from a total of 226 pixels, with 224 pixels exhibiting a FWHM of below 100 eV at 97 keV. While this result represents excellent fabrication yield, the dominant failure modes of the missing 30 pixels were the separation of the bulk absorber from the sensor or the rupturing of the thin TES membrane. Figure 2 depicts the excellent uniformity for the resolution of individual pixels of the LANL-NIST microcalorimeter array where the majority of pixels achieve a FWHM between 50 and 60 eV at 97 keV. A spectrum composed of the energy calibrated data of the 226 individual detectors is shown in the inset of Figure 2. This spectrum illustrates the 97 and 103 keV photopeaks associated with the decay of ^{153}Gd . The FWHM of the 97 keV ^{153}Gd photopeak in the summed spectrum results was determined to be 59.4 eV.

Measurements of mixed-isotope Pu standards

Measurements of the X - and γ -ray emission spectra from a series of mixed-isotope Pu materials were performed to characterize the performance of the

Figure 2: FWHM of the 97 keV photopeak of a ^{153}Gd check source for individual pixels of the LANL-NIST γ -ray spectrometer. The majority of the 226 pixels displayed achieve a FWHM of 50-60 eV at 97 keV. The spectrum depicting the 97 and 103 keV photopeaks of ^{153}Gd is shown in the inset. This spectrum is the sum of the data from 226 energy calibrated individual pixels of the LANL-NIST γ -ray spectrometer.

160 LANL-NIST γ -ray spectrometer. This work represents the first time the si-
 161 multaneous readout of all 256 pixels for high-resolution measurements of these
 162 material types has been achieved. The high-quality spectra obtained from these
 163 measurements, shown in Figure 3, were used to determine the relative abun-
 164 dances of Pu isotopes present in each sample. The samples studied in this work,
 165 listed in Table 1, span a range of Pu isotopic content corresponding to a variety
 166 of isotopic enrichment values. The Pu isotopic content of each source, corrected
 167 for the time since fabrication, is also included in Table 1. The total Pu mass
 168 of each of the CBNM samples measured for this work was approximately 5.5 g
 169 while the Pu mass of the FZC-158 source was approximately 0.7 g, all in a
 170 Pu-oxide form. The most abundant isotopic constituents of the CBNM sources
 171 are ^{239}Pu and ^{240}Pu with the bulk of the material being composed of the for-
 172 mer. The FZC-158 source differs from the CBNM sources in that its ^{239}Pu
 173 content is relatively low and ^{240}Pu accounts for 94% of the total Pu mass. The

174 decrease in the relative ^{239}Pu content from CBNM-PU70 to CBNM-PU93 asso-
 175 ciated with higher enrichment samples is easily observed in the high-resolution
 176 spectra of Figure 3. As the relative ^{239}Pu content decreases, the peak heights of
 177 the 129.296 and 203.55 keV photopeaks associated with the decay of ^{239}Pu also
 178 decrease relative to the multitude of other transitions observed in each spec-
 179 trum. The ultra-high energy resolution capabilities of the LANL-NIST γ -ray
 180 spectrometer allow for the observation of the increase in the ^{240}Pu relative abun-
 181 dance compared to that of ^{239}Pu for the FZC-158 standard even in the complex
 182 100 keV region, shown in Figure 4. The 104.234 keV photopeak correspond-
 183 ing to ^{240}Pu decay becomes more prominent as the relative mass abundance of
 184 ^{240}Pu increases in FZC-158 compared to the CBNM-type sources.

185 Total data acquisition times for each sample range from a total of 78.5 h for
 186 the Pu isotopic standard FZC-158 to 95.4 h for CBNM-PU70. Total event rates
 187 were typically limited to 1-3 events per second per pixel, corresponding to array
 188 event rates of up to 600 cps. Approximately 2 mm of Cd shielding was used to
 189 attenuate the prevalent 59.9 keV ^{241}Am transition for the CBNM type materials.
 190 Due to the relatively low photon emission rate of the FZC-158 standard, no
 191 Cd shielding was required to obtain single pixel count rates of approximately
 192 0.5 cps. For the measurements of these Pu isotopic standards, spectroscopic data
 193 obtained from approximately 195 pixels was used to create the total summed
 194 spectra shown in Figure 3. The majority of the omitted data from operating
 195 pixels was due to difficulties in maintaining SQUID lock for incident photon
 196 energies greater than 150 keV despite exhibiting excellent energy resolution. By
 197 removing single pixel data in which there exists a discrete cutoff at a particular
 198 energy we avoid the introduction of potential discontinuities in the detection
 199 efficiency curve used to extract the relative Pu mass abundances.

200 The relative Pu isotopic abundances determined from the summed spectra
 201 are presented in Table 1. The photopeak areas corresponding to transitions
 202 following the decay of specific Pu isotopes, normalized by the known emission
 203 probability of each observed transition, were used to deduce the relative Pu
 204 isotopic abundances [13]. The uncertainty associated with each reported rel-

205 active Pu isotopic abundance includes only contributions from statistical error,
 206 dominated by the number of counts in each photopeak and the peak fitting pro-
 207 cedure used to determine peak area. It should be noted that recent studies have
 208 established that the total measurement uncertainties associated with the deter-
 209 mination of relative Pu mass abundances is limited by the precision to which
 210 the constants of nature used in the calculation (e.g. emission probabilities of
 211 individual photons, decay half-lives of Pu isotopes, etc.) are known. A detailed
 212 discussion and comparison of these uncertainty calculations can be found in the
 213 forthcoming work of Hoover *et al.* [14].

214 Inspection of the reported and measured Pu mass abundance ratios listed
 215 in Table 1 indicates very good agreement between the expected isotopic ratios
 216 and the measurements of the LANL-NIST γ -ray spectrometer. In general, for
 217 the CBNM type isotopic standards, the expected relative mass abundances are
 218 within measurement uncertainty of the values deduced from the series of mea-
 219 surements in this work. However, the spectral analysis of the FZC-158 source
 220 results in significant deviations from the reported isotopic compositions, par-
 221 ticularly for the $^{238}\text{Pu}/^{239}\text{Pu}$ and $^{241}\text{Pu}/^{239}\text{Pu}$ relative isotopic abundances.
 222 The decay time-corrected values for the $^{238}\text{Pu}/^{239}\text{Pu}$ and $^{241}\text{Pu}/^{239}\text{Pu}$ isotopic
 223 ratios are 1.28 and 13.5%, respectively. These values are at variance to the mea-
 224 surement results of 0.77(2) and 7.2(2)% for the $^{238}\text{Pu}/^{239}\text{Pu}$ and $^{241}\text{Pu}/^{239}\text{Pu}$
 225 isotopic ratios. For this source, the values reported in Table 1 are obtained from
 226 the HPGe spectra analyzed with the FRAM software package [15]. These HPGe
 227 studies of the FZC-158 source suggest that the reported isotopic composition of
 228 the FZC-158 source may not be accurate [16]. Further studies of the isotopic
 229 content of the FZC-158 standard are necessary to understand the discrepancies
 230 between the results of this work and the reported Pu isotopic composition of
 231 this source.

232 A slight degradation of the average array energy resolution is observed when
 233 measuring the complex photon emission spectrum of the mixed-isotope Pu stan-
 234 dards compared to that of the ^{153}Gd check source. The individual pixel resolu-
 235 tions of the 102.98 keV ^{241}Am photopeak for a single 22 h measurement period

Figure 3: Observed X- and γ -ray emission from 55 to 215 keV for the Pu isotopic standards CBNM-PU70, CBNM-PU84, CBNM-PU93, and FCZ-158. For these energy-calibrated summed spectra the FWHM of the 102.98 keV ^{241}Am transition was determined by a Gaussian fit to be 70.3, 82.0, 79.7, and 91.0 eV for FCZ-158, CBNM-PU70, CBNM-PU84, and CBNM-PU93, respectively.

Figure 4: (Color online) An overlay of the observed X- and γ -ray emission from 96 to 105 keV for the Pu isotopic standards CBNM-PU70 (solid black), CBNM-PU84 (dashed blue), CBNM-PU93 (dotted red), and FZC-158 (solid black with diagonal fill). The prominent photopeaks have been labeled. Events corresponding to the escape of a Sn X-ray prior to the complete thermalization of the energy of an incident photon are observed at 100.81, 100.852, and 104.15 keV.

236 of CBNM-PU84 at approximately 1.5 cps/pixel is shown as the red dashed line
 237 histogram in Figure 5. The average measured FWHM of the individual pixels
 238 of the LANL-NIST γ -ray spectrometer is 72.1 eV for these data. This is an
 239 increase from the average individual pixel FWHM of the 103.18 keV photopeak
 240 from a ^{153}Gd check source using identical array operational parameters and
 241 nearly identical total array count rates, shown as the black solid line histogram
 242 in Figure 5. Under identical operating conditions, the average individual pixel
 243 FWHM for a ^{153}Gd source at 103.18 keV was determined to be 64.1 eV. The
 244 degraded average pixel resolution is most likely the consequence of an increased
 245 frequency of SQUID unlock events in the presence of a significantly larger flux
 246 of high energy γ -rays emitted from the mixed-isotope Pu materials.

Pu Standard	$^{238}\text{Pu}/^{239}\text{Pu}$ (%)	$^{240}\text{Pu}/^{239}\text{Pu}$ (%)	$^{241}\text{Pu}/^{239}\text{Pu}$ (%)	$^{241}\text{Am}/^{239}\text{Pu}$ (%)
CBNM-PU93	0.0102	6.74	0.0645	0.278
	0.0099(2)	6.7(1)	0.0657(7)	0.282(5)
CBNM-PU70	0.94	24.9	2.05	6.84
	0.96(1)	25.1(3)	2.05(2)	7.00(9)
CBNM-PU84	0.0673	16.81	0.3263	1.12
	0.0654(7)	16.42(3)	0.3266(2)	1.12(1)
FZC-158	0.83(1)	8600(100)	7.6(1)	54.7(5)
	0.77(2)	8000(200)	7.2(2)	51(1)

Table 1: The mixed-isotope Pu standards and their relative isotopic abundances studied with the LANL-NIST γ -ray spectrometer. The top value for each isotopic ratio entry corresponds to the expected Pu composition of the listed isotopic standard, corrected for the time since fabrication. The bottom value for each isotopic ratio entry is extracted from data obtained from measurements using the 256-pixel microcalorimeter array. The top entries for the FZC-158 Pu material are values determined by the FRAM analysis of HPGe spectra.

Analysis of multi-channel, high-resolution data

While the energy resolution of individual microcalorimeter detectors is extraordinary, the ultimate goal is to obtain a single spectrum of similar spectroscopic quality for each measurement that is to be used for the isotopic analysis of the studied material. Ideally, the summed spectrum resolution would not be significantly degraded compared to that of single pixel spectra. However, the process of summing individual energy-calibrated spectra becomes non-trivial for the ultra-high energy resolutions achieved by the microcalorimeter detectors. Issues in energy calibration of the intrinsically non-linear microcalorimeter detectors were described by Hoteling *et al.* [17]. In a many-pixel array containing detectors with energy resolutions of 80 eV at 100 keV, offsets in the energy calibration of individual spectra as small as 50 eV were shown to produce noticeably distorted peak shapes. Therefore, the accuracy of the energy calibration of individual spectra must be lower than 0.02% to minimize peak distortions in the summed spectra of high-resolution data. As will be shown in the following, this requirement is difficult to meet with traditional high-order polynomial fits to a selection of observed transitions spanning the dynamic range of the microcalorimeter detectors.

Figure 5: (Color online) Comparison of individual pixel resolutions for a measurement of the photon emission spectra of the CBNM-PU84 and ^{153}Gd sources under identical array operating conditions. The black solid line histogram depicts the FWHM of the 103.18 keV photopeak of the ^{153}Gd check source with an average value of 64.1 eV. The red dashed line histogram shows the individual pixel FWHM at the 102.98 keV photopeak associated with the decay of ^{241}Am with an average value of 72.1 eV.

Manual data processing of large amounts of multi-channel data quickly becomes intractable. To minimize the time required to analyze a data set, an automated data analysis toolkit was developed in the ROOT framework [18]. Prior to the energy calibration, the pretrigger baseline, baseline RMS, and pulse position within the digital record of each current pulse are deduced for each event. Recorded events for which these observables are beyond a 3-sigma range of the average value for a data set are rejected. Pile-up events are also rejected. To minimize the contribution of stationary noise to the pulse height determination, optimal filtering is performed on each pulse record of the remaining events [19].

Typical data acquisition time intervals range from 16 to 21 h and are limited by the hold time of the cryostat and ADR. During this measurement time, slight temperature-dependent gain drifts occur for each individual detector. While a number of drift correction techniques are available, the variable span Super-Smoother algorithm of J.H. Friedman *et al.* [20] is both effective and reliable for the automated processing of many single pixel spectra. Currently, the drift cor-

rection offset applied to single pixel data is energy independent and is deduced
 from a single strong transition near the center of the dynamic range of each
 detector. A non-linear energy calibration is then applied to the drift-corrected
 single pixel spectra. Depending on the isotopic content of the mixed-isotope
 Pu material, nine to thirteen reference transitions are selected as calibration
 points. These calibration points typically span an energy range from the 59.9
 to 208 keV transitions associated with the decay of ^{241}Am . If a 7th-order poly-
 nomial is used to fit these reference points, the deviations of the centroids of
 the peaks in the spectrum from the actual energy of each transition are still
 large enough to degrade the resolution and cause peak shape abnormalities in
 a summed spectrum containing many channels of data. A natural cubic spline
 calibration is employed to minimize these deviations. Figure 6 depicts the en-
 ergy offsets of the centroids a calibrated single pixel spectrum to the energy
 values used for the calibration. The ten reference energy points used for this
 calibration were fit with 6th- and 7th-order polynomial functions as well as a
 cubic spline function. As shown in the bottom portion of Figure 6, the energy
 offset, even for reference energies used in the calibration, can differ from the
 actual energy value by up to 20 eV for both the 6th- and 7th-order polynomial
 fit to the data. The offsets generated using a cubic spline function to fit the ten
 reference energy points are significantly improved and are within 5 eV of the
 calibration energy values. For single pixel energy resolutions of 50 to 60 eV, a
 20 eV offset in the position of the energy calibrated peak can cause significant
 degradation of the energy resolution in a summed spectrum containing many
 individually calibrated single pixel spectra. For reference, the top portion of
 Figure 6 shows the cubic spline fit to the ten reference energy points used for
 calibration and the typical linearity of an individual detector. The differences
 between the cubic spline fit and 6th- and 7th-order polynomial fits to these data
 points are not discernible in this figure.

The improvement in the accuracy of the energy calibration using a cubic
 spline fit function is best illustrated by examining the calibrated position of peak
 centroids that are not used as reference transitions in the energy calibration.

Figure 6: (Color online) A cubic spline fit of ten reference energy points from single pixel CBNM-PU70 data (top portion) and the energy calibration offsets for 6th- and 7th-order polynomial and the cubic spline fit to these reference energy points (bottom). The dotted blue, dashed red, and solid black lines represent the offsets of the 6th- and 7th-order polynomial and cubic spline fits, respectively. Energy offsets from the energy reference points of up to 20 eV for the polynomial type fits are observed. The energy offsets are all below 3 eV from the energy reference points using the cubic spline calibration function.

311 The red open triangle markers of Figure 7 represent the centroid position of
 312 the 104.234 keV photopeak corresponding to the decay of ^{240}Pu using a 7th-
 313 order polynomial function for energy calibration following drift correction for
 314 each pixel of the LANL-NIST γ -ray spectrometer. There remain up to 60 eV
 315 offsets from the centroid of the calibrated single pixel spectra to the expected
 316 energy using this calibration function. Using a cubic spline function for energy

317 calibration, the majority of centroids of the calibrated spectra are within 10 eV
 318 of the expected value as shown by the black square markers of Figure 7. The
 319 decreased spread in the values of the calibrated peak centroids results in an
 320 improved energy resolution as well as the mitigation of peak shape deformation
 321 when many calibrated spectra from individual detectors are summed to produce
 322 a final spectrum.

Figure 7: (Color online) The centroid of the Gaussian fit to the 104.234 keV ^{240}Pu photopeak for individual pixels of the LANL-NIST γ -ray spectrometer. The red open triangle markers depict the centroid position of the 104.234 keV photopeak following the energy calibration using a 7th-order polynomial fit of ten reference energies. The black square markers correspond to a cubic spline function fit of the same data during energy calibration. The majority of the energy offsets using the cubic spline calibration function are within 10 eV of the expected value while energy offsets of up to 60 eV are observed using the polynomial fit.

323 For transitions in spectral regions where there are few reference point op-
 324 tions, the cubic spline calibration results in a significantly improved resolution
 325 and peak shape for the summed spectrum. The bottom portion of Figure 8
 326 depicts the summed peak shape of the 203.55 keV transition associated to the
 327 decay of ^{239}Pu . This photopeak is typically too weak for use as a reference point
 328 for the energy calibration of individual pixels. The dashed blue and dotted red
 329 spectra represent CBNM-PU70 summed data using the 6th- and 7th-order poly-
 330 nomial energy calibrations, respectively. The FWHM of 180.8 eV of this peak
 331 using the 6th-order polynomial energy calibration is an improvement compared

332 to that of the 7th-order polynomial, 234.5 eV. There also exists a significant
 333 low energy tail when using the 7th-order polynomial as an energy calibration
 334 function. Both the peak shape, energy resolution, and energy centroid of the
 335 CBNM-PU70 summed spectrum are further improved by the cubic spline en-
 336 ergy calibration, shown as the solid black spectrum in Figure 8. A FWHM of
 337 125.2 eV for the 203.55 keV photopeak is achieved using a cubic spline energy
 338 calibration.

339 The summing of data from multiple pixels with different resolutions intro-
 340 duces non-Gaussian peak shapes in the final summed spectrum. These arti-
 341 facts can not be corrected for with the previously presented energy calibration
 342 and drift correction techniques. Figure 9 shows an overlay of three individual
 343 drift-corrected and energy calibrated CBNM-PU84 single pixel spectra. The
 344 102.98 keV and 104.234 keV photopeaks of ²⁴¹Am and ²⁴⁰Pu as well as the
 345 103.74 keV Pu K_{α1} X-ray are displayed. When the individual pixel spectra
 346 are summed, the differences in energy resolutions will result in the development
 347 of small high- and low-energy photopeak tails. This is generally a small effect
 348 on the overall peak shape of the summed spectrum but needs to be taken into
 349 account during the peak fitting procedure for the accurate determination of the
 350 above-background peak areas, particularly in regions of significant peak overlap.

351 High event rate data acquisition

352 High-statistics spectra are vital to the measurement quality for nuclear safe-
 353 guards applications. For this reason, the maximum count rate of microcalorime-
 354 ter arrays must be significantly increased to be comparable to those achieved
 355 by HPGe detectors. Aspects previously prohibiting the simultaneous readout
 356 of 256 multiplexed pixels for Pu isotopic measurements included data through-
 357 put limits of the computer interface card, the performance of software drivers,
 358 and the ability to maintain an array-wide SQUID lock. These experimental
 359 hurdles have been overcome in this work. For the time-domain multiplexing of
 360 array signals, the sampling rate of each pixel limits the application of the digital

Figure 8: (Color online) The spectra in the top portion of this figure represent the 100 keV region of the CBNM-PU70 summed pixel data using 7th-order polynomial (dotted red) and cubic spline (solid black) energy calibration functions. There is a slight improvement in energy resolution in this energy region when using the cubic spline function compared to the polynomial fit. The bottom portion of the figure contains spectra depicting the significant improvement in energy resolution, peak centroid location, and peak shape observed when using the cubic spline function in spectral regions with few energy calibration points. The spectrum produced using a 6th-order polynomial energy calibration is represented as a dashed blue line. For the 203.55 keV photopeak of ²³⁹Pu, FWHM of 180.8, 234.5 and 125.2 eV are achieved using the 6th- and 7th-order polynomials and cubic spline energy calibration functions, respectively.

361 feedback required to maintain SQUID lock. Incident high energy photons can
 362 cause a change in the error signal that requires a feedback signal larger than
 363 the digital feedback circuitry can provide. In these cases, SQUID lock will be
 364 lost and a relock procedure initiated. While the relatively low energy photons

Figure 9: (Color online) An overlay of three single pixel CBNM-PU84 drift corrected and energy calibrated spectra. The 102.98 keV and 104.234 keV photopeaks of ^{241}Am and ^{240}Pu are shown. The 103.74 keV photopeak corresponds to the Pu $K_{\alpha 1}$ X-ray. The observed variation of resolutions of the single pixel spectra result in the development of small tails on both the high- and low-energy side of the summed photopeak.

365 emitted from a ^{153}Gd source do not cause a large number of SQUID unlock
 366 events, the frequency of these events increases with thermalization of high en-
 367 ergy gamma-rays emitted by a mixed-isotope Pu material and can quickly cause
 368 array operational instability. In previous generations of microcalorimeter arrays,
 369 this problem was mitigated by decreasing the number of rows to be multiplexed
 370 and thus increasing the sampling rate. Until recently, software monitoring tools
 371 were used to inspect the array for unlocked SQUIDs and, if necessary, initiate a
 372 SQUID relock procedure. These tools were effective at maintaining SQUID lock
 373 across the array for low-energy photon measurements and high energy photon
 374 measurements with relatively low count rate. With increased flux of high en-
 375 ergy photons, the speed of the SQUID relocking software becomes insufficient
 376 to maintain the array lock stability. The ability to maintain array-wide SQUID
 377 lock for large energy incident photons has been dramatically improved with
 378 the introduction of the latest generation of digital feedback electronics. The
 379 strict control of the latency of the applied feedback signal from the measured
 380 error signal as well as the migration from software-based control to firmware-

381 based digital feedback card relock functionality have significantly improved the
 382 SQUID-lock stability of the array during operation. These advances not only
 383 improve the current array stability during measurements of mixed-isotope Pu
 384 samples but also dramatically improve operational stability and performance
 385 for array count rates beyond 1 kHz.

Figure 10: (Color online) Comparison of individual pixel FWHM at 103 keV for ^{153}Gd (solid black) and CBNM-PU70 (dashed red) at total raw array count rates of 1.25 kHz. The average pixel FWHM were determined to be 72.2 and 92.1 eV for ^{153}Gd and CBNM-PU70, respectively. The resolution degradation observed in this subset of pixels is representative of the behavior of the entire array.

386 Figure 10 depicts the single pixel FWHM of the 103 keV photopeak for the
 387 ^{153}Gd check source and CBNM-PU70 isotopic standard for total array count
 388 rates beyond 1 kHz. The data were obtained at total raw array event rates
 389 of 1.25 kHz, approximately 5.6 cps/pixel, during fully multiplexed, 8x32 mea-
 390 surements. At present, typically 20% of the data are rejected during offline
 391 processing when omitting pulse records that contain pileup events. The average
 392 pixel FWHM for the ^{153}Gd data is 72.2 eV at this high count rate. Though
 393 its a measured increase from the value of 64.1 eV previously reported for lower
 394 count rate measurements, the array resolution performance is excellent for total
 395 event rates at and beyond 1 kHz. The resolution of the individual pixels during
 396 the CBNM-PU70 measurement is degraded compared to the ^{153}Gd measure-

397 ments due to the higher frequency of SQUID unlock events resulting from the
 398 thermalization of relatively large energy photons. The increased SQUID un-
 399 lock frequency also has a deleterious effect on peak shapes in the CBNM-PU70
 400 energy calibrated summed spectrum. Figure 11 shows the 100 keV energy re-
 401 gion of the CBNM-PU70 summed spectrum for both low and high event rate
 402 measurements. The FWHM of the 102.98 keV ^{241}Am photopeak was measured
 403 to be 92.1 eV at 1.25 kHz. The significant low- and high-energy tails of the
 404 photopeaks in this spectrum are a consequence of similar peak shapes for in-
 405 dividual pixel spectra due to the increased frequency of SQUID unlock events.
 406 It should be noted that the data analysis techniques presented in this article
 407 may not be optimal for the analysis of high count rate data. The development
 408 of analysis techniques for high count rate microcalorimeter data has recently
 409 been addressed by Alpert *et al.* [19] and the application of these techniques is
 410 underway.

Figure 11: (Color online) Overlay of the energy calibrated CBNM-PU70 summed spectrum for 1.25 kHz total array count rate (dotted red) and a low event rate measurement (solid black). The FWHM of the 102.98 keV ^{241}Am photopeak was determined to be 92.1 eV at 1.25 kHz. Noticeable low- and high-energy tails are observed for the photopeaks shown in this spectrum due to the higher frequency of SQUID unlock events for this high rate mixed-isotope Pu standard measurement.

Conclusions and Future Work

This article reports the first simultaneous readout of all 256 pixels of the LANL-NIST γ -ray spectrometer for measurements of mixed-isotope Pu isotopic standards. High precision measurements of the Pu isotopic composition of the standards CBNM-PU70, CBNM-PU84, CBNM-PU93, and FZC-158 have been performed. The LANL-NIST γ -ray spectrometer exhibited excellent energy resolution during full array measurements with the average individual pixel FWHM typically between 70 and 80 eV. Regarding the analysis of individual pixel spectra, utilizing a cubic spline fit function for energy calibration and the Super-Smoother algorithm of J.H. Friedman[20] for the gain-drift correction of single pixel data were shown to be effective in maintaining excellent energy resolution and minimizing anomalous peak shapes when adding a large number of channels to obtain a final summed spectrum. The extracted peak areas of the summed spectra were used to deduce Pu isotopic ratios in very good agreement with the reported values for the CBNM type Pu isotopic standards. The understanding of systematic errors is an area of current and future work and the study of the contribution of aspects such as peak shape models, efficiency curve models, and the fitting of non-Gaussian peak shapes is underway.

The LANL-NIST γ -ray spectrometer has demonstrated stable operation and obtained high-resolution data at total event rates beyond 1 kHz. For the measurement of the CBNM-PU70 standard at a total event rate of 1.25 kHz, the average pixel FWHM was 83.2 eV for the 103 keV photopeak of ^{241}Am . Additional digital feedback and room temperature electronics upgrades are planned for the near future which will further improve the stability and resolution performance of this novel instrument for high-count rate studies of complex X - and γ -ray emission. The application of recent advances in the analysis of high-count rate data, specifically the ability to extract pulse height information from pulse records that contain pile-up events while retaining the ultra-high resolution performance of the LANL-NIST γ -ray spectrometer, will effectively increase the maximum achievable total event rate for this microcalorimeter array. These

441 studies, as well as the push for the stable operation at even higher total event
 442 rates, are topics of present and future work.

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