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Abstract

Superconducting Gamma-ray detectors offer an order of magnitude higher energy resolution than conventional high-purity germanium detectors. This can significantly increase the precision of non-destructive isotope analysis for nuclear samples where line overlap affects the errors of the measurement. We have developed Gamma-detectors based on superconducting molybdenum-copper sensors and bulk tin absorbers for nuclear science and national security applications. They have, depending on design, an energy resolution between ~50 and ~150 eV FWHM at ~100 keV. Here we apply this detector technology to the measurement of uranium isotope ratios, and discuss the trade-offs between energy resolution and quantum efficiency involved in detector design.

Introduction

Gamma spectroscopy is widely used to determine the isotopic composition of nuclear materials[1]. Upon decay, each isotope emits Gamma rays and X-rays with characteristic energies, and their intensity can be related to the isotope abundance. High-precision isotope ratio measurements are typically based on emission lines with similar energies to reduce errors due to the energy dependence of self-absorption and detection efficiency. These lines often overlap with each other when analyzed with a conventional

high-purity germanium (HPGe) spectrometer. This increases the statistical errors, and thus reduces the limiting precision of the measurement.

One important example is the measurement of uranium enrichment. For routine analysis, NaI detectors are typically used to infer the level of enrichment from the strength of the 186 keV line of U-235 above the Compton background. If higher precision is required, HPGe detectors can be used to infer enrichment from the emission of the thorium daughters at 92 keV, since U-238 decays into Th-234 with Gamma emission lines at 92.4 and 92.8 keV, and the nearby Th $K_{\alpha 1}$ X-ray at 93.35 keV originates mostly from the decay of U-235[2,3,4]. This approach works well with HPGe detectors for moderate levels of enrichment when the characteristic emission lines have roughly equal strength. Line overlap makes this approach less precise in cases of very high or very low levels of enrichment, when weak emission lines in close vicinity to strong ones have to be analyzed.

Superconducting Gamma-spectrometers can address the line overlap problem, since their energy resolution is not limited by the statistics of electron-hole pair generation. They can reduce statistical errors, and can thus make the 92 keV region of the Gamma-spectrum more relevant for measuring uranium enrichment. Here we discuss the performance of superconducting Gamma-spectrometers, their relevance for measuring uranium enrichment, and the trade-offs involved in the detector design.

Spectrometer

Cryogenic gamma-ray microcalorimeters consist of a gamma-ray absorber attached to a highly sensitive thermometer. The thermometer often consists of a thin film

superconductor, also known as a transition-edge sensor (TES), operated in the narrow temperature range of the transition from its superconducting to normal state. Here its resistance changes rapidly as a function of temperature so that the energy of a single gamma-photon can be determined with extremely high precision. The absorber and the thermometer are strongly thermally coupled together, while they are weakly coupled to the cold bath. In the simplest case[5], random thermal fluctuations across this weak thermal conductance G set the limit on the energy resolution of microcalorimeters to $\Delta E_{FHWM} \approx 2.355\zeta\sqrt{k_B T^2 C_{abs}}$, where, ζ is a factor of order 1-2 that depends on the thermal properties of the thermometer, T is the absolute temperature and C_{abs} is the heat capacity of the absorber. For high energy resolution, the operating temperature T and the heat capacity C_{abs} of the microcalorimeter must therefore be as small as feasible. For practical applications, an operating temperature of 0.1K and absorber volume ~ mm^3 offer a compromise between ease of operation and performance. This produces, depending on material choices and absorber sizes, cryogenic spectrometers with energy resolution between 50-150 eV FWHM with quantum efficiency around 50% at 100 keV.

At LLNL, we are developing cryogenic gamma ray spectrometers based on bulk Sn absorber and Mo/Cu multilayer transition-edge sensors[6, 7,8]. The TES thermistors are made photolithographically on 4" Si wafers, and a ~ mm^3 Sn absorber is glued to each sensor with stycast epoxy. The spectrometer is cooled to its operating temperature of 0.1K in a two-stage adiabatic demagnetization refrigerator (ADR)[9,10]. The detector is voltage biased at the onset of its superconducting-to-normal transition and exposed to radiation. The current signal from the spectrometer is readout by a low noise dc superconducting quantum interference device (SQUID) current amplifier. The gamma

induced pulse signals are further amplified at room temperature and digitized by a 14-bit data acquisition system. The data are subsequently optimally filtered to extract spectral information.

Simulations

A Monte-Carlo N-Particle (MCNP) simulation was performed to estimate the performance of the cryogenic gamma-ray spectrometer in measuring ^{235}U enrichment. The response of a cryogenic detector with a 1 mm^3 Sn-absorber and with an energy resolution of 100 eV FWHM was compared to that of an HPGe detector with an energy resolution of 500 eV FWHM for a natural uranium sample (figure 1). The ^{234}Th gamma lines at 92.38 keV and 92.80 keV and the Th $K\alpha_1$ x-ray at 93.35 keV are fully resolved by the cryogenic spectrometer with 100 eV FWHM, while they blend together when using an HPGe detector. This illustrates the potential of high-resolution cryogenic spectrometers to determine isotope ratios, provided the total number of counts is sufficient for good statistics.

To optimize the detector design and trade-offs in energy resolution and quantum efficiency, we can quantify the statistical error in measuring enrichment as a function of energy resolution. For this, we consider the case when two emission lines with intensities I_1 and I_2 at energies E_1 and E_2 are examined with a spectrometer with an energy resolution ΔE_{FWHM}. We assume that the spectrometer response can be described by a Gaussian function and that the Compton background B is constant over the energy range of interest. In this case, the statistical error σ_1/I_1 measurement can be calculated analytically[10,11]. Figure 2 shows this limiting error as a function of detector resolution ΔE_{FWHM} for a given line separation $E_1 - E_2 = 550$eV and $I_1/I_2 = 1\%$ for different number of

total counts $I_{total} = I_1 + I_2$. As expected, the percent statistical error decreases with decreasing ΔE_{FWHM} rather sharply as long as there is line overlap, but then levels off as the detector resolution is sufficient to fully separate the lines of interest, with the remaining reduction in error being due to a better discrimination of the signal from the Compton background B. We see that a detector with energy resolution of ~ 300 eV FWHM is already sufficient to fully resolve the Th Kα_1 line at $E_1 = 93.35$ keV from the ^{234}Th gamma line at $E_2 = 92.8$ keV, at which point the precision of the enrichment measurement then depends mostly on the total number of counts and approaches $\sigma_1 \approx \sqrt{I_1}$ as expected.

Results

We have measured a weakly radioactive (20 nCi) low-enriched uranium sample to evaluate the performance of our cryogenic gamma ray spectrometer to determine ^{235}U enrichment using Th Kα_1 at 93.35 keV as a measure of ^{235}U and the ^{234}Th gamma lines at 92.38 and 92.80 keV as a measure of ^{238}U (figure 3). This particular cryogenic detector has an energy resolution of 90 eV FWHM and can thus well resolve the relevant gamma and x-ray lines. Also, the Compton background count is low and the assumption that the detector response can be characterized by a Gaussian function is justified. For comparison, we include a measurement of the same sample with a planar HPGe spectrometer, taken over the same acquisition time of ~ 3 days as the data from the single pixel cryogenic spectrometer.

The isotopic abundances of ^{235}U (A_{235}) and ^{238}U (A_{238}) are extracted from the observed peak intensities I_1 (Th Kα_1) and I_2 (^{234}Th) by $\frac{A_{235}}{A_{238}} = \frac{I_1}{I_2}\frac{T_1}{T_2}\frac{\eta_2}{\eta_1}\frac{B_2}{B_1}$, where $T_{1,2}$ are

the half-lives, $\eta_{1,2}$ are detection efficiencies and $B_{1,2}$ are the branching ratios. Since the two lines are closely spaced in energy, we can assume $\eta_1/\eta_2 = 1$. Note that the literature values for branching ratios for the gamma and x-ray lines in the 90-100 keV region of U spectrum quoted in the literature vary significantly[4]. In this paper we use 0.0260, 0.02560 and 0.0550 as the branching ratios for the emission lines at 92.38, 92.790 keV and 93.356 keV, respectively, since these values are used by the U235 analysis code developed at LLNL for measuring uranium enrichment[3].

We determine the ^{235}U enrichment of this sample to be (1.14 ± 0.14) %. For comparison, the enrichment value extracted with the U235 code from the HPGe spectrum is (1.017 ± 0.014) %. This indicates that a single pixel cryogenic gamma spectrometer is not sufficient to improve the precision of the measurement, because of its small detection efficiency, despite the exquisitely high-energy resolution. This is due to the fact that the line overlap problem of HPGe detector is compensated for by the increased detector efficiency they offer, especially, when analyzing weak radioactive samples as in this experiment.

Discussion

The low number of counts in the weak 93.35 keV x-ray line limits the precision of ^{235}U enrichment measurement for a single pixel cryogenic spectrometer. However, the statistical error can be reduced by increasing absorber volume thus the quantum efficiency of each pixel and by fabricating large detector arrays. For example, for uranium analysis, increasing the size of each individual absorber by a factor of 10 could increase the number of counts. Although this will degrade energy resolution by a factor

of ~ $\sqrt{10}$, it will improve precision of enrichment measurement as long as there is no line overlap (cf. figure2).

In addition, a spectrometer composed of a 100-pixel array, where each pixel retains the high energy resolution will increase the total number of count by hundred fold and reduce the statistical error by a factor of ~ 10. We are currently building such a multi-pixel cryogenic spectrometer[12]. It will reduce the statistical error by a factor of ~30 and improve the statistical precision of enrichment measurement to ~ 0.001%.

Conclusions

Cryogenic Gamma-ray spectrometers offer higher energy resolution than conventional HPGe spectrometer. This can, for example, improve the measurement of uranium using the 92 keV region, provided the total numbers of counts in the lines of interest are sufficient. We have built a superconducting gamma ray spectrometer based on Sn absorbers and Mo/Cu multilayer sensors with an energy resolution of 90 eV FWHM at ~ 100keV. We have used it to analyze a low-enriched uranium sample and find for this application, the highest signal-to-noise ratio is achieved with energy resolution 300 eV FWHM. Note that for other applications such as Pu isotope analysis much higher energy resolution is needed[13]. Larger absorber sizes and arrays of 100 pixels are needed to improve the precision of the measurement to ~ 0.001%.

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List of Figures:

Figure 1: MCNP simulations of an HPGe spectrometer with 500 eV FWHM and a cryogenic spectrometer with 100 eV FWHM. The geometry used in the simulation does not take into account all surrounding materials and thus underestimates the Compton background. Note the increased number of escape lines due to the small pixel size of cryogenic detector. However, since these lines can be fully resolved, they do not affect the precision of the measurement.

Figure 2: Statistical error for measuring a line at energy E_1 with intensity I_1 in the presence of a much stronger line at E_2 with intensity $I_2 = 100\ I_1$ as a function of detector energy resolution ΔE_{FWHM}. The simulation assumes an energy separation of E_1-E_2 = 550 eV, corresponding to the separation of ^{234}Th line at 92.80 keV and the Th Kα_1 at 93.35 keV, and a constant Compton background B. As expected, the relative statistical error decreases with improved ΔE_{FWHM} rather sharply as long as there is line overlap, but then levels off as the detector resolution is sufficient to fully separate the lines of interest, with the remaining reduction in error being due to a better discrimination of the signal from the Compton background B. For well-separated lines the precision of the enrichment measurement depends mostly on the total number of counts and approaches $\sigma_1 \approx \sqrt{I_1}$.

Figure 3: Gamma spectra of a low-enriched uranium sample taken with a cryogenic spectrometer with an energy resolution of 90 eV FWHM and a planar HPGe spectrometer with an energy resolution of 600 eV FWHM. The emission lines at 92.38 keV and 92.80 keV and at 93.35 keV, which are a measure of ^{238}U and ^{235}U concentration respectively, are fully resolved by the cryogenic detector.

Figure 1

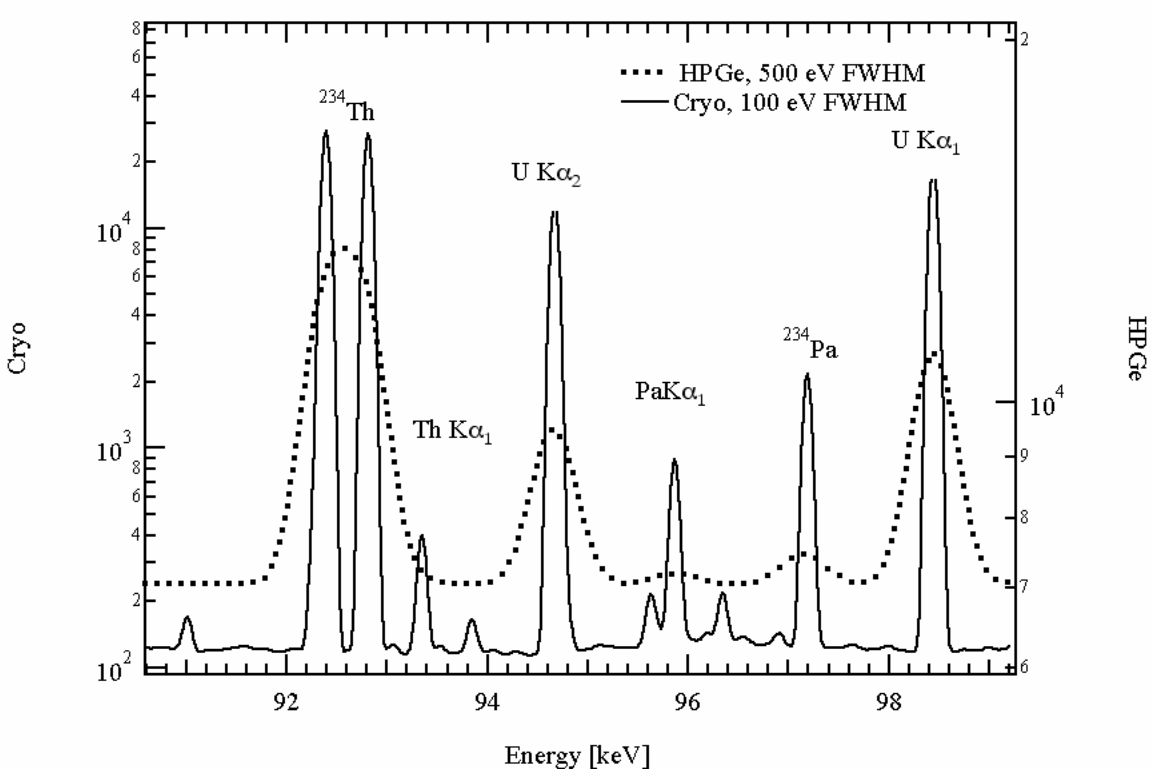

HPGe, 500 eV FWHM
Cryo, 100 eV FWHM
^{234}Th
U Kα$_2$
U Kα$_1$
Th Kα$_1$
PaKα$_1$
^{234}Pa
Cryo
HPGe
Energy [keV]
92
94
96
98

Figure 2

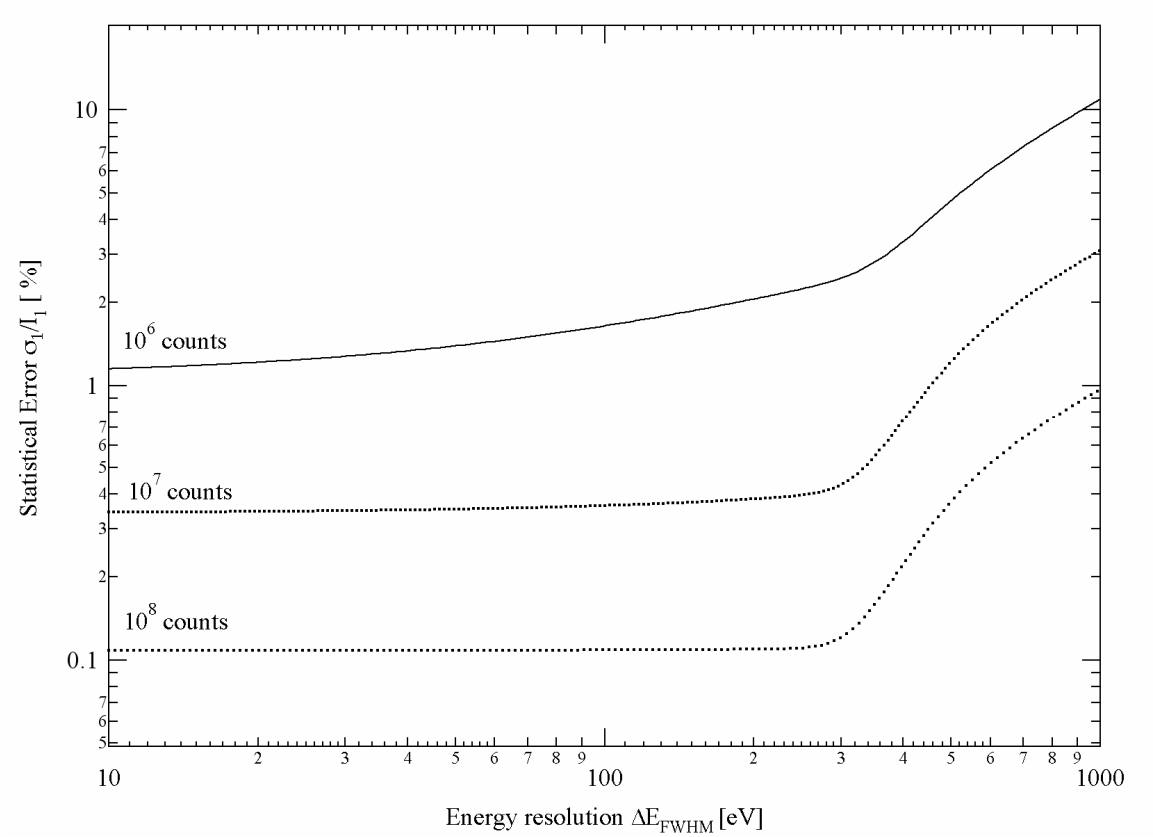
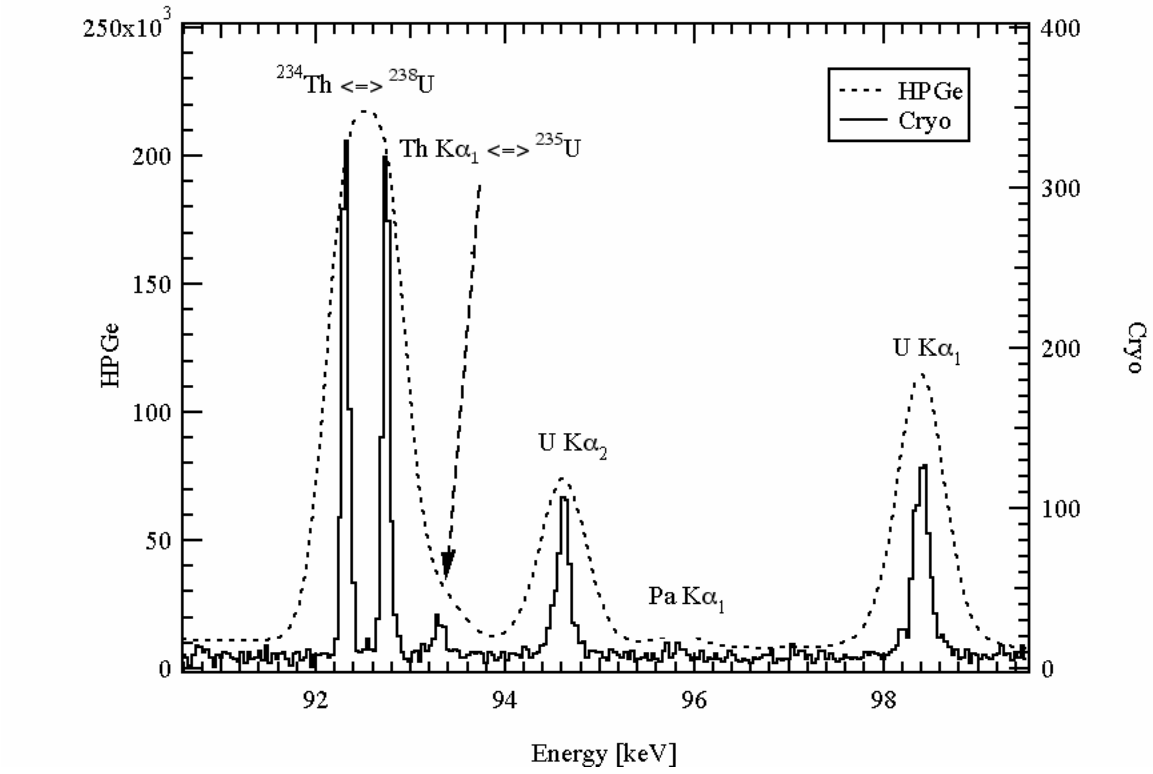

Statistical Error σ_1/I_1 [%]
10
1
0.1
10^6 counts
10^7 counts
10^8 counts
10
100
1000
Energy resolution ΔE_{FWHM} [eV]

Figure 3


250x10^3
400
234Th <=> 238U
HPGe
Cryo
Th Kα1 <=> 235U
200
300
150
U Kα1
HPGe
Cryo
200
100
U Kα2
50
100
Pa Kα1
0
0
92
94
96
98
Energy [keV]