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Analysis of Nuclear Material by Alpha Spectroscopy with a Transition-Edge Microcalorimeter

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Abstract We present measurements from a cryogenic microcalorimeter designed to detect alpha particles. The enhanced resolution of microcalorimeter alpha detectors will provide new capabilities for actinide analysis. We demonstrate a spectral resolution of 2.4 keV full width at half maximum (FWHM) for 5.3 MeV alpha particles from a ^{210}Po source. In addition, we present an alpha spectrum from ^{209}Po showing the first direct measurement of decay into the ^{205}Pb ground state. Finally, measurements of 100 keV gamma-rays from a Gd source show an ultimate alpha particle resolution of 159 eV to be achievable which may provide an avenue for investigating ion energy loss mechanisms in bulk materials.

Keywords Microcalorimeter · Alpha spectroscopy · Transition edge sensor · Nuclear forensics

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1 Introduction

Microcalorimeters operate on the principle that any energy that can be converted into phonon excitations or heat, can be detected with a thermometer. By using very sensitive thermal detectors, cryogenic microcalorimeters have already shown excellent resolution for the detection of single photons with wavelengths ranging from γ-rays to near-infrared [1]. However, this technology is not exclusive to the sensing of photons. In this paper, we present results for the use of microcalorimeters as high resolution alpha particle detectors with applications for the nuclear forensics community.

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Alpha particle spectroscopy is the preferred method for analyzing radioactive materials in trace quantities. For these small samples, alpha particle emission dwarfs the probability of a γ-ray decay. One example is ^{239}Pu, where alpha decay is 2000 times more likely than γ decay [2].

High resolution alpha particle spectra are normally taken using Passive Implanted Planar Silicon (PIPS) or other Si based detectors. The energy resolution of these detectors is typically 10–30 keV, with better resolution obtained at the expense of collection area and count rate. In addition, the resolution of Si detectors is fundamentally limited to ~ 8.5 keV at 5.5 MeV by factors which include fluctuations in energy lost to a surface dead layer, fluctuations in energy available for ionization, fluctuations in created electron-hole pairs, and charge collection efficiency [3]. An additional factor obscuring accurate results is straggle, which is alpha particle energy lost when passing through a material. Si detectors suffer straggle from both the thickness of the emitting source and an unavoidable surface dead layer on the detector.

The resolution limits of the Si detector are serious detriments for its use in nuclear forensics. When a mixture of radioactive actinides must be assayed, overlap of elements necessitates chemical separation of each element, which is a labor intensive process. Additionally, even after chemical separation, some very important isotopic ratios are still indistinguishable. One such example is in the determination of the ratio of ^{240}Pu/^{239}Pu. The relative abundance of ^{240}Pu allows the determination of the history and intended use of a nuclear reactor from which the sample was taken [4]. However, the most intense emission lines from these isotopes are only 12.9 keV apart. Another important assay is the detection of naturally occurring and fissionable ^{235}U, which is made difficult by the proximity of its emission line to ^{236}U. The ^{235}U/^{236}U ratio is a characterization of the processing history of a radioactive sample [5]. Furthermore, the presence of ^{236}U in the environment is a means of separating contamination effects due to nuclear reactors from naturally occurring uranium deposits [6]. Despite these closely spaced emission lines, quantitative analysis is still possible with Si detectors in the presence of this overlap using deconvolution algorithms. However, large errors are introduced and the required counting time is long [7, 8]. As a result, time intensive and costly mass spectroscopy is necessary to determine the ^{240}Pu/^{239}Pu and ^{235}U/^{236}U isotopic ratios.

Cryogenic microcalorimeters should provide the increase in resolution necessary to analyze mixed actinide and closely spaced isotopic ratios without the need for chemical separation or mass spectroscopy, thereby increasing forensic throughput. The fundamental resolution limits of microcalorimeters have been discussed [1, 9], and as an example, our group has already shown resolutions in detecting gamma-rays at 100 keV of 25–50 eV full width at half maximum (FWHM). The energy resolution of a microcalorimeter scales as the square root of the highest detectable energy, so for alpha particles at 5 MeV, one would expect a resolution of 180–375 eV FWHM.

We present in this manuscript the design of a cryogenic microcalorimeter for alpha particle detection. Alpha particles with energy 5.3 MeV from ^{210}Po were detected with resolution of 2.4 keV, a factor of 3–4 better than the limit of Si detectors. Another type of microcalorimeter, published by Leblanc et al., with a germanium thermometer coupled to a copper and germanium absorber has obtained 3.5 keV FWHM resolution [10]. With the improvement in resolution shown by the TES microcalorimeter,

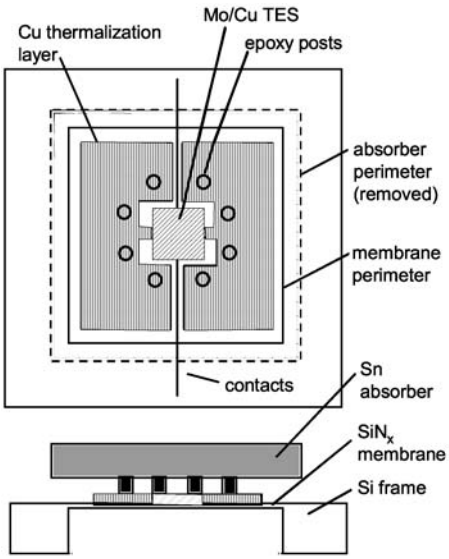

Fig. 1 Schematic top (*upper*) and side (*lower*) views of the microcalorimeter alpha detector. Schematic is not to scale. For the device described here, the Sn absorber is 1.7 mm × 1.7 mm × 0.25 mm, and the Mo/Cu TES is 0.4 mm × 0.4 mm × 0.25 μm

the first observation of an alpha particle emission from the ^{209}Po ground state is presented. Additionally, photons from a gamma-ray source at 100 keV confirm that at low energies, the resolution of the alpha particle detector is 170 eV FWHM, as expected from thermal properties and signal to noise measurements. The discrepancy between expected and observed resolutions may be attributable to fluctuations in lattice damage caused by the alpha particles.

2 Device Design

Microcalorimeters consist of two elements, a bulk absorber to convert incident energy into phonons or heat, and a thermometer to measure the change in temperature. The absorber is superconducting Sn which has low heat capacity, thereby increasing the signal to noise response to an incident alpha particle. Also, Sn has well characterized and adequate thermalization properties, increasing allowable particle count rate [11, 12]. The thermometer is a Transition-Edge Sensor (TES) which is a thin film superconductor electrically biased in the superconducting-to-normal transition so that a small change in temperature yields a large change in resistance. The TES in use at NIST is comprised of a bilayer of molybdenum and copper with a tunable transition temperature near 0.1 K [13].

A schematic of the alpha particle microcalorimeter is shown in Fig. 1. The superconducting Sn absorber is positioned atop eight 20 μm tall posts made from photoimagable epoxy. The posts attach to a thin-film Cu thermalization layer that connects to the TES via two narrow fingers. There are two reasons for the use of a Cu thermalization layer. First, the heat capacity provided by the additional Cu allows for high energy particles to be absorbed without saturating the superconducting transition of a standard TES. In fact, the TES used for this detector is identical to ones

used for both X- and γ-ray detectors whose properties are well understood [6–8]. The second motivation for the thermalization layer is that it allows eight posts to be located off the TES, thereby reducing stress on the thermometer film. The eight posts provide mechanical stability for a large area absorber to be attached, allowing more collection area, while eliminating position dependence from alpha strikes on various parts of the absorber. The TES and thermalization layer are collocated on a suspended silicon nitride membrane to provide thermal isolation from the Si frame.

3 Measurements

The TES of the microcalorimeter alpha particle detector is electrically biased into its superconducting transition, so it is necessary to cool the detector below the transition temperature of ~ 0.1 K. Therefore, the detectors are mounted in a two stage adiabatic demagnetization refrigerator capable of cooling to 0.055 K. An alpha source of interest is mounted on the same stage as the detector negating the need for infrared filters, and reducing source straggle.

We have performed initial measurements on a mixture of ^{210}Po and ^{209}Po. The polonium mixture is electroplated onto a platinum disk which is mounted facing the microcalorimeter detector. Electroplating is a useful process for creating thin samples and thus minimizing straggle. The ^{210}Po provides a single alpha emission at 5305 keV without interference from any nearby alpha decay. The results from the microcalorimeter measurements as well as data from a quality commercial Si detector are shown in black in Fig. 2. The data are fitted by a convolution of a left handed exponential decay, to simulate the source straggle, with a Gaussian to emulate the detector resolution. The fits are shown in red and are given by

$$f(E) = \frac{A\mathrm{Exp}[\frac{\sigma^2+2s(E-E_0)}{2s^2}]\mathrm{Erfc}[\frac{\sigma^2+s(E-E_0)}{\sqrt{2}s\sigma}]}{2s}, \tag{1}$$

where Erfc is the complementary error function, the peak area is A, σ is the detector resolution, s is the width of the straggle, and E_0 is the location of the peak. The results yield a FWHM resolution for the microcalorimeter alpha detector of 2.42 ± 0.07 keV, as compared to the Si detector with a resolution of 7.4 ± 0.2 keV. In addition, the microcalorimeter straggle has a width of 1.75 ± 0.05 keV due to source thickness with no additional straggle contributed by the detector. The Si detector, however, yields a straggle width of 4.7 ± 0.2 keV due to both the source and the surface dead layer of the Si.

With the excellent resolution of the microcalorimeter, we are able to make the first measurement of the branching ratio of ^{209}Po alpha decay into the ground state of Pb and its first excited state. ^{209}Po will decay with 0.48% probability into a nuclear spin state of ^{205}Pb that is 262.8 keV above its ground state and 99.3% probability into either the ground state or the first excited state 2.3 keV above [14]. The existence of the two lowest states is known from gamma ray emission from the 262.8 keV state into the lowest two. However, the resolution limitations of Si alpha detectors have never allowed a measurement of the alpha decay branching ratio into the lowest two

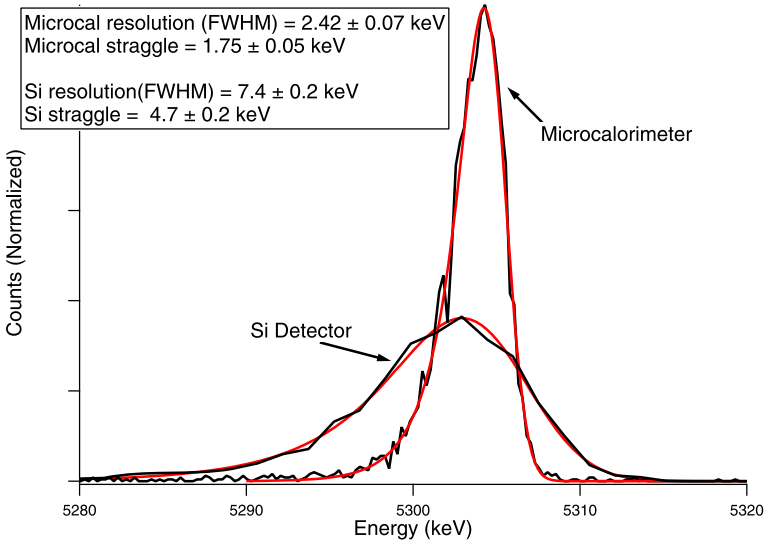

Fig. 2 (Color online) Spectrum of ^{210}Po alpha emission (*black*) taken by a microcalorimeter alpha particle detector and PIPS detector. The fits (*red*) are given by (1) and yield the parameters shown in the plot

states [10]. Figure 3 shows the ^{209}Po alpha emission where the tallest peak is the first excited state, 2.3 keV above the ground state, and the ground state is a high energy shoulder in the plot. The data are fitted with (1) summed for two peaks, and the ground state decay probability is $25 \pm 6\%$ based on peak areas.

The 2.4 keV resolution of the microcalorimeter alpha detector is worse than the resolution expected from studies of previous gamma-ray detectors, approximately 180–350 eV FWHM. In fact, we measured the alpha detector's response to low energy (100 keV) photons from a Gd gamma-ray source. The resulting spectrum is shown in Fig. 4. The pulse height response with respect to incident energy is almost linear from the gamma-ray energies of 100 keV up to the alpha particle energies of 5.5 MeV, with a response degradation $\approx 1.5\%$ at higher energies. Given the linearity of the detector response, the same resolution is expected for incident alpha particles. The resolution in response to the low energy photons is 159 eV FWHM. Another measure of the expected resolution is the prediction of the optimal filter given the average alpha particle signal and the average baseline noise. This measure yields 150 eV FWHM. We conclude that there exists a mechanism for energy fluctuations in the system when alpha particles are incident. The only possible scenarios seem to be sputtering, fluorescence escape, and lattice damage. Fluorescence escape is not observed in the TES detector, therefore, this system may provide a means for the characterization of the energetics of lattice damage by single ions.

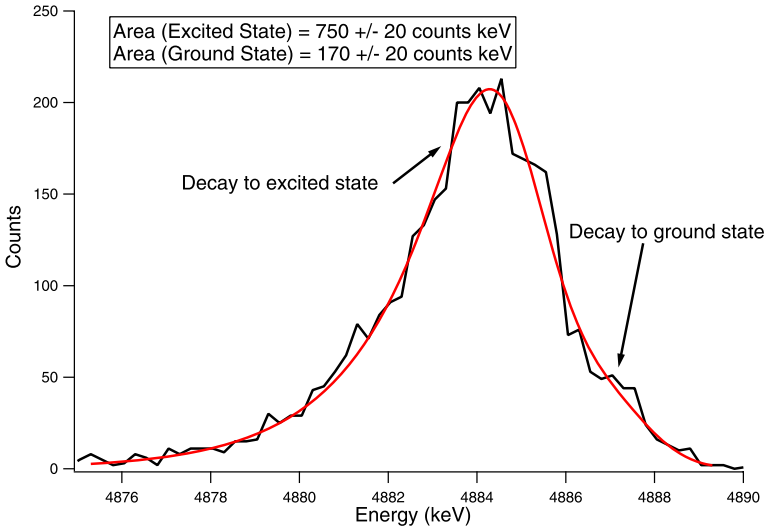

Fig. 3 (Color online) Microcalorimeter alpha particle spectrum (*black*) of ^{209}Po. Two alpha emission peaks are present: decay to the first excited state of ^{205}Pb and decay to the ground state which is 2.3 keV more energetic. *The red curve* is a fit with a sum of two peaks described by (1) and yielding the fit parameters shown in the plot

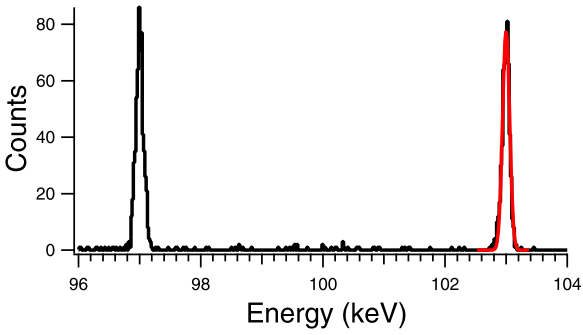

Fig. 4 (Color online) Spectrum (*black*) of pulses from the microcalorimeter alpha detector in response to gamma-rays at 97 and 103 keV. The resolution (FWHM) from a Gaussian fit (*red*) is 159 eV

4 Conclusions

Microcalorimeters provide a substantial benefit to the detection of alpha particles which will have a large impact on the nuclear forensics community. We have demonstrated a resolution of 2.4 keV FWHM at 5.3 MeV with the measurement of a ^{210}Po source. This is a factor of 3–10 improvement over the current Si detector technology. With this increase in resolution, the ground state decay of ^{209}Po has been observed. Based on gamma-ray measurements, resolutions as good as 159 eV are expected for alpha particles. The discrepancy between the predicted and observed resolution is an indication of a separate energy loss mechanism. This may provide a route toward understanding the energies of ion damage in bulk materials.

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