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Toward a 256-Pixel Array of Gamma-Ray Microcalorimeters for Nuclear-Materials Analysis

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Abstract We are developing a gamma-ray spectrometer for the analysis of nuclear materials based on an array of superconducting transition-edge-sensor microcalorimeters. The instrument will include eight columns of time-division-SQUID multiplexing circuitry capable of reading out 256 sensors. Our most recent sensors are bulk (1.5 mm square $\times$ 0.25 mm thick) superconducting Sn absorbers glued to Mo/Cu bilayer thermometers. When fully populated, the active area of the spectrometer will be 5.76 cm^2, and the maximum count rate of the array will approach 20 kHz. Thus, our spectrometer will be comparable to the state-of-the-art 100 keV high-purity-Ge detector in count rate and collecting area, but with an order of magnitude better energy resolution. Half the detectors will be optimized for operation up to 100 keV, and the other half for operation up to 200 keV. A version of the spectrometer with a partially populated detector array was delivered to Los Alamos National Laboratory in June, 2007. We describe the present status of that instrument. In addition, we review results from a prototype array of 14 detectors that achieved 47 eV average energy resolution (full width at half maximum at 103 keV) and 25 eV resolution in the best detector. An important application of this technology is determining the total

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Pu content in spent reactor fuel without detailed knowledge of the reactor's operating history.

Keywords Gamma-ray spectrometer · Microcalorimeter · Transition-edge sensor

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Gamma-ray spectroscopy is an important technique in the nondestructive assay of nuclear materials. The state-of-the-art spectrometer is the high-purity, planar Ge detector, which has a collecting area of a few cm^2, a photon count rate of ≈ 50 kHz, and energy resolution of ≈ 500 eV (full width at half maximum, or FWHM) when measuring 100 keV photons. Recently, several groups have demonstrated microcalorimeter spectrometers with sub-100 eV energy resolution near 100 keV [1–4], which is good enough to separate most lines in the complicated 100 keV γ-ray spectra of nuclear materials. Because individual γ-ray microcalorimeters are relatively small (≈ 1 mm^2) and slow (50–100 Hz), a useful instrument will require an array of microcalorimeters to increase the collecting area and count rate. In this paper, we first review the performance of our prototype, 14-pixel, multiplexed array of 100 keV transition-edge-sensor (TES) microcalorimeters. Next, we introduce our 256-pixel architecture and describe the construction and testing of the first instrument based on that architecture. Finally, we discuss some future steps needed for a complete laboratory instrument.

A TES microcalorimeter uses a thin film, electrically biased into its superconducting-to-normal-metal transition, as a sensitive thermometer. Deposited heat from an absorbed photon increases the temperature and resistance of the film. The resulting pulsed decrease in the device current is used to measure the energy of the photon.

The 14-pixel prototype array is pictured in Fig. 1. The individual detectors are similar to the one presented by Zink et al. [2]. Each TES is a 400 μm square thin-film bilayer of Mo and Cu with a superconducting transition temperature tuned to $T_c \approx$ 100 mK and a normal-state resistance of $R_n \approx 8$ mΩ. Cu bars abate "unexplained noise" [5] and tune the transition width [6]. A 6.25 mm chip contains a 4×4 array of detectors on a 1.3 mm pitch. In order to stop the γ-rays, a bulk, polycrystalline-Sn absorber (900 μm square × 250 μm thick) is glued to a lithographically patterned epoxy post on each detector. The calculated quantum efficiency of the absorbers is about 25% for 100 keV photons.

The 14-pixel array was read out with two SQUID amplifier columns by use of a cryogenic time-division multiplexer (TDM) [7–9]. The 2-column × 8-row TDM had two unused rows in column 2. Because the TDM architecture was developed for arrays of high-speed (sub-100 μs) X-ray microcalorimeters [10], reading out 14 relatively slow γ-ray TESs, which had an average pulse-decay time constant of 2 ms, was not difficult. The performance of the detectors was not measurably degraded by the multiplexed readout. Based on previous work [10], we calculate that the TDM system would require an open-loop bandwidth of $f_{OL} \geq 2$ MHz and nonmultiplexed amplifier noise (referred to the first-stage SQUID) of $N_{SQ1} \leq 0.3\ \mu\Phi_0/\sqrt{\mathrm{Hz}}$ in order to read out an 8-column × 32-row array of 25 eV-resolution γ-ray microcalorimeters

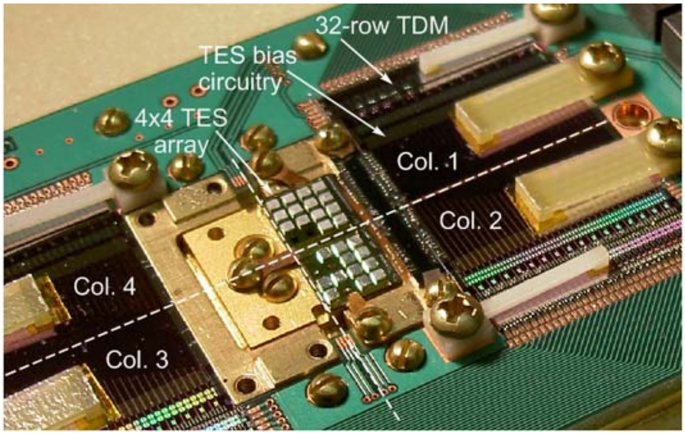

Fig. 1 (Color online) Two 4 × 4-pixel detector array chips and supporting cryogenic electronics. To build each array chip, the Sn absorbers were assembled into a micromachined caddy, and then all 16 were glued to the array at once. Of the 32 possible detectors on the two chips, 14 survived absorber assembly and cooldown. The interface-wiring and SQUID-TDM chips shown here contain circuitry to read out 128 detectors through four amplifier columns. The entire detector plane is cooled to 50 mK by an adiabatic-demagnetization refrigerator

with a degradation in resolution of no more than 5%. The TDM system used in the 14-pixel experiments, which was not near the calculated performance limits, approached these requirements, with $f_{\mathrm{OL}} = 1.6$ MHz and $N_{\mathrm{SQ1}} = 0.47\mu\Phi_0/\sqrt{\mathrm{Hz}}$.

To measure its spectroscopic performance, the 14-pixel array was illuminated by a ^{153}Gd γ-ray source. The count rate recorded by each detector was 7 Hz. After pileup events were removed, pulses due to γ-rays were optimally filtered [11]. The resulting pulse-height spectrum from each detector was spline corrected to remove gain drifts over time and then converted to energy units by use of a linear fit to the known 97.43 and 103.18 keV γ-ray lines. The energy resolution of the best pixel was $\Delta E_{\mathrm{FWHM}} = 25$ eV at 103 keV (see Fig. 2). The weighted-average energy resolution of the 14 pixels was $\langle\Delta E_{\mathrm{FWHM}}\rangle = 47$ eV at 103 keV, meaning that a measurement made with the array would attain the same statistical significance per unit time as would one made with a 14-pixel array of identical, 47 eV-resolution detectors. The 14-pixel experiment is described in much greater detail by Doriese et al. [3].

Based on the results from the 14-pixel prototype array, we set out to design a 256-pixel instrument, including a new unit pixel, a new detector-array chip, and a new cryogenic platform. The new unit pixel (see Fig. 3a) moves the absorber-support structures off the TES. Our testing of similar devices has yielded evidence that supporting the absorber with posts and glue placed directly on the TES can distort the superconducting transition shape unpredictably. Whether this effect is related to mechanical stresses by the glue on the detector membrane, a chemical interaction between the TES bilayer and some component of the glue, or some other effect remains unclear. In the new device, copper bars thermally connect the absorber-support posts to the TES.

Our new array chip (Fig. 3b) contains 66 pixels, half of which are optimized for sensitivity to 100 keV photons, and half for 200 keV photons. The unusual shape of

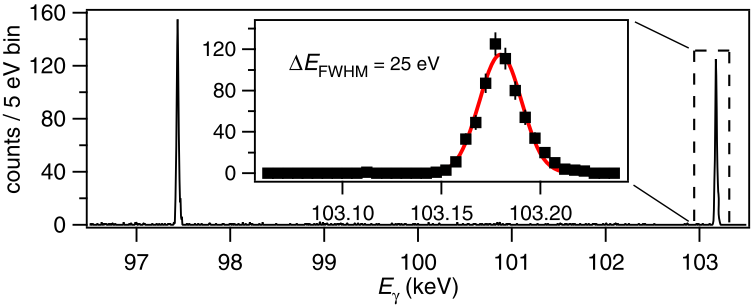

Fig. 2 (Color online) The best performance of a single detector in the 14-pixel prototype array, $\Delta E_{\text{FWHM}} = 25$ eV. This performance was repeatable over months, and was achieved both with the 2×8 TDM and in single-detector mode

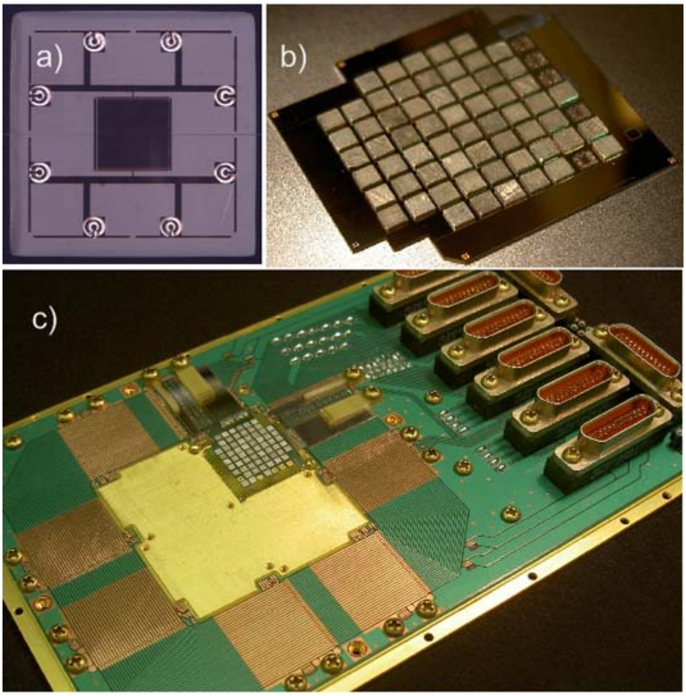

Fig. 3 (Color online) (**a**) Top view of a new detector before the Sn absorber is attached. The absorber would be glued to the eight epoxy posts (clear C's). Heat is channeled to the TES through Cu bars (*dark lines*). This entire structure is suspended on the membrane, which extends to the dark region at the edges of the image. In this pixel, which is intended for 200 keV operation, the TES (*center, striped square*) is 400 μm on a side. A second design (not pictured) optimized for 100 keV photons is similar, but has a 250 μm-square TES. (**b**) An array chip of 66 TES's, of which 61 have a 1.5 mm-square Sn absorber. Half the pixels are optimized for 100 keV operation, and half for 200 keV. (**c**) The 256-pixel detector plane, with one quadrant (one detector chip and superconducting integrated circuitry for two columns of the 8-column $\times$ 32-row SQUID TDM) populated

the chip outline is to allow four chips to be tiled into a 45 mm-square detector plane, while being held down with Be-Cu spring clips and no glue. Au wirebonds thermally sink the array chips to the bulk-Cu substrate. Up to 64 pixels can be read out from each of the four chips, by use of an 8-column × 32-row SQUID TDM. Figure 3c shows the 256-pixel detector plane, with one quadrant populated.

A requirement for a fieldable nuclear-materials-analysis instrument is that its cryogenic system must be easy to operate. To this end, we designed a new cryogenic platform consisting of a modified NIST adiabatic demagnetization refrigerator [12] (ADR) mounted on a commercial 3 K pulse-tube cryocooler in a compact physical arrangement. The pulse tube eliminates the expense and complication of liquid cryogens. The cryogenic system performed well; in initial tests with no detector wiring or other nonparasitic load, the ADR held for more than 7 days below 100 mK. With the full cryogenic wiring for the 256-pixel array installed, and one quadrant operating, the ADR held for 24 hours below 65 mK and 48 hours below 100 mK. A γ-ray TES operating in the electrical environment of the pulse tube showed no degradation in performance. We have yet to test the full system with operational detectors.

The instrument was delivered to a nuclear-materials-analysis laboratory at Los Alamos in June, 2007. The initial array included with this instrument (pictured in Fig. 3c) suffered from a variety of mechanical and chemical problems due to improper mixing of the glue during the absorber attachment. Among other problems, the epoxy catalyst attacked the Mo in the detector bias wires, causing a variety of open-circuit and weak-link phenomena. Although the pixels were not useful as γ-ray spectrometers, we were able to use the TDM readout electronics to measure the critical temperatures of about 40 TESs. The measured T_cs fell in a band of 106–110 mK, in line with the design values. New detector arrays are underway, and will be installed in the LANL instrument soon.

An important upgrade to the system will be real-time data-analysis software that will demultiplex the raw data stream, trigger, filter pulses, and calibrate energy. Work on such a software package, based on algorithms developed by NASA-Goddard for the Astro-E and Astro-E2 satellite experiments [13], is nearing completion.

To conclude, we have designed a 256-pixel γ-ray spectrometer for nuclear-materials analysis, based on a 14-pixel prototype array. The prototype array achieved 25 eV energy resolution at 103 keV in the best pixel, and 47 eV average energy resolution over the 14 pixels. The new 256-pixel instrument, designed with ease of use in mind, was operated at Los Alamos in June, 2007 with a partially filled detector array. When fully populated with detectors, this spectrometer will be a unique tool for the analysis of nuclear materials, combining the superior energy resolution of microcalorimeters with the large count rate and collecting area that are so valuable in HPGe detectors.

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