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A Liquid-Cryogen-Free Cryostat for Ultrahigh Resolution Gamma-Ray Spectrometers

Jonathan G. Dreyer, Theo Hertrich, Owen B. Drury, Jens Höhne, Stephan Friedrich

Abstract— We are developing ultra-high energy resolution gamma-ray detectors based on superconducting transition edge sensors (TESs) for nuclear non-proliferation and fundamental science applications. They use bulk tin absorbers attached to molybdenum-copper multilayer TESs, and have achieved an energy resolution between 50 and 90 eV FWHM for gamma-ray energies below 122 keV. For increased user-friendliness, we have built a cryostat that attains the required detector operating temperature of 0.1 K at the push of a button without the use of cryogenic liquids. It uses a two-stage mechanical pulse tube refrigerator for precooling to ~3 K, and a two-stage adiabatic demagnetization refrigerator for cooling to the base temperature. The cryostat is fully automated, attains a base temperature below 30 mK without the use of cryogenic liquids, and has a hold time of ~2 days at 0.1 K between 1-hour demagnetization cycles. Here we discuss the performance of the cryostat for operation in a Gamma-spectrometer with 112-pixel arrays of superconducting TES detectors.

Index Terms—Microcalorimeters, Mo/Cu multilayers, detector arrays, ultra-high resolution gamma ray spectroscopy, pulse tube refrigerator, adiabatic demagnetization refrigerators

I. INTRODUCTION

GAMMA (γ) spectrometry is one of several analytical techniques used in the characterization of nuclear material. Characteristic γ -rays, unique to each radioisotope, provide a signature from which the composition of the sample can be determined. Relative line intensities from these radioisotopes are used to determine isotope ratios and infer sample age, origin and processing history. High-purity germanium (HPGe) detectors are often used for γ -ray analysis, since they provide high energy resolution for separating the emission from different isotopes and high absorption efficiency necessary to measure weak lines from dilute samples. Isotopic ratios can be measured with HPGe detectors with an error of ~1%, limited either by statistical errors of the counting statistics, or by systematic errors in detection

efficiency, and background subtraction. To limit the systematic errors due to the uncertainty in detection efficiency, intense γ -lines with similar energies, typically around ~100 keV, are measured. Unfortunately, γ -ray spectra in this energy range are frequently affected by line overlap when using HPGe detectors, introducing uncertainties into the measurement. The statistical errors from this line overlap can be reduced by the use of ultra-high energy resolution γ -spectrometers that can fully separate the emission lines from each other. This separation also reduces the systematic errors from background subtraction, and allows more precise detection efficiency corrections.

Cryogenic γ -spectrometers based on superconducting microcalorimeters operated at temperatures of ~0.1 K offer an order of magnitude improvement in energy resolution over conventional HPGe spectrometers (figure 1) [1]. Our microcalorimeters consist of bulk superconducting Sn absorbers coupled to sensitive Mo/Cu superconducting-to-normal transition edge sensors (TESs), both weakly coupled to a cold bath [2 - 4]. Ultra-high energy resolution requires operating temperatures of 0.1 K, a regime that many users are not experienced with. For increased user-friendliness, we have built a cryostat that is fully automated and operates the microcalorimeters at temperatures of 0.1 K without the use of liquid cryogens. It uses a mechanical pulse tube cryocooler and a two-stage adiabatic demagnetization refrigerator. Here we describe design of the cryostat and its performance.

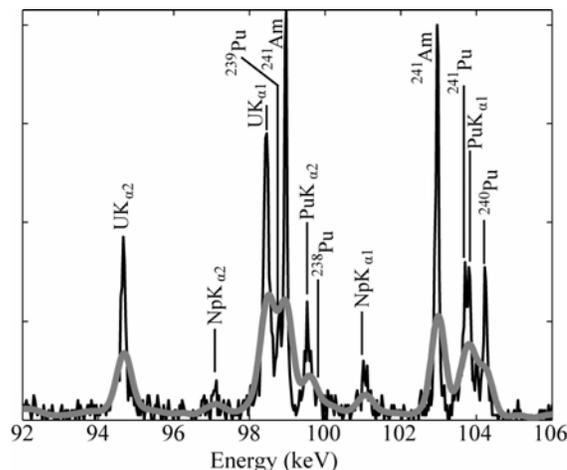


Fig. 1. Spectrum of a mixed isotope Pu sample from a microcalorimeter spectrometer (thin black line), compared to the idealized Fano-limited response of a HPGe detector (thick grey line). The energy resolution of the microcalorimeter detector allows separating the U, Np, Pu and Am lines, which remain affected by line overlap even for an ideal HPGe detector.

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II. CRYOGENIC GAMMA RAY MICROCALORIMETERS

Microcalorimeter detectors consist of a radiation absorber, a sensitive thermometer, and a weak thermal link to the cryostat cold bath. Our γ -ray microcalorimeter detectors use bulk tin absorbers with dimensions between $\sim 1 \times 1 \times 0.25 \text{ mm}^3$ and $\sim 2 \times 2 \times 0.5 \text{ mm}^3$. They are attached to Mo/Cu multilayer thermometers on a silicon nitride (SiN) membrane that forms the weak thermal link to the cold bath. Incident γ -radiation with energy E_γ interacts within the Sn absorber with heat capacity C , and the absorbed energy increases the temperature by $\Delta T \approx E_\gamma/C$. This change in temperature is measured with a Mo/Cu multilayer thermometer operated in the steep transition between its superconducting and its normal state (figure 2), as the signal ΔT is proportional to the absorbed energy E_γ . Random thermal fluctuations $4k_B T^2 G$ across the SiN weak link with thermal conductance G fundamentally limit the energy resolution ΔE_{FWHM} of the γ -ray microcalorimeter to

$$\Delta E_{\text{FWHM}} \approx 2.355 \sqrt{4k_B T^2 C}. \quad (1)$$

This energy resolution can be well below 100 eV FWHM for $T \approx 0.1 \text{ K}$ and heat capacities C of a few pJ/K [3, 5].

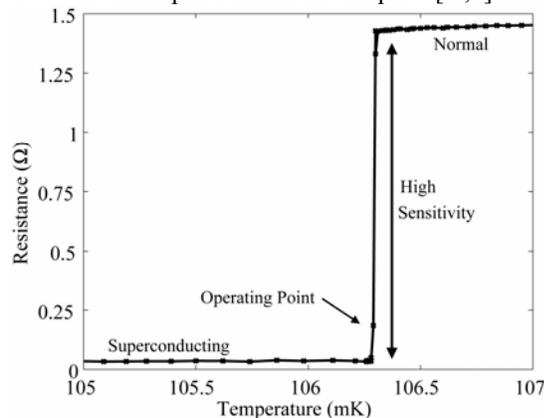


Fig. 2. Superconducting-to-normal transition in a Mo/Cu multilayer TES microcalorimeter. The sharp transition at the operating point ensures high sensitivity.

Since the volume of individual sensors is limited to $\sim \text{mm}^3$, and the $\sim \text{ms}$ decay time constants prevent data acquisition at high rates, we and several other groups are developing pixilated detector arrays with multiplexed readout for increased sensitivity [4] [5]. The cryostat discussed here will initially house 112-pixel detector arrays, but its $4'' \times 4''$ detector stage is laid out for substantially larger arrays in future spectrometer upgrades.

III. CRYOSTAT DESIGN

Superconducting X-ray detectors are becoming an increasingly mature technology, but their operation is currently limited to a few research groups with low-temperature expertise. To broaden the use of these detectors in nuclear applications, we have built a cryostat that attains the required operating temperatures below $\sim 0.1 \text{ K}$ at the push of a button without the use of cryogenic liquids. The instrument was designed in collaboration between the Advanced Detector

Group at Lawrence Livermore National Laboratory and VeriCold Technologies, and was built by VeriCold (figure 3). Like earlier cryogenic microcalorimeter X-ray spectrometers, it uses a two-stage pulse-tube refrigerator for precooling to $\sim 3 \text{ K}$, and a two-stage adiabatic demagnetization refrigerator for cooling to its base temperature below 0.1 K [6]. The design incorporates several elements from earlier γ -ray spectrometers based on liquid N_2 and He precooling that have been in operation at Lawrence Livermore National Laboratory since 1999 [3, 4]. It improves upon that design by removing the need for cryogenic liquids, and by increasing reliability and hold time at low temperature.



Fig. 3. Photograph of the γ -ray spectrometer. The detectors are held on a short cold finger behind the thin round Be window in the front, and the preamplifier electronics is held in a shielded enclosure on the opposite side of the cryostat. The cylinder on top of the cryostat contains the gas reservoir of the pulse tube refrigerator, and compressor and rotary valve are separated from the cryostat body to reduce vibrations.

A. Two-stage pulse tube refrigerator

Precooling to a temperature of $\sim 3 \text{ K}$ is achieved with a two-stage pulse tube refrigerator (PTR) [7, 8]. Like Stirling or Gifford-McMahon cryocoolers, pulse tube refrigerators use the compression and expansion of He gas to achieve cooling. The main advantage of PTRs is that their mechanically vibrating parts such as the rotary valve can be separated from the cold head. This greatly reduces the vibrations at the low-temperature detector stage and the associated microphonic noise. The main disadvantage of PTRs is their comparably low efficiency, which makes them less attractive for space-based applications, but matters little for laboratory based nuclear forensics and non-proliferation applications.

The compressor produces periodic gas pressure variations at a frequency of a few Hz set by the rotary valve. Pulse tube refrigerators consist of two columns, one of them (the “pulse tube”) being empty, and one of them (the “regenerator”) filled with porous solids (figure 4). In the regenerator and in the heat exchangers on top and at the bottom of the pulse tube, the gas is thermally well coupled to its environment, while the gas inside the pulse tube is thermally isolated and its temperature therefore varies with pressure.

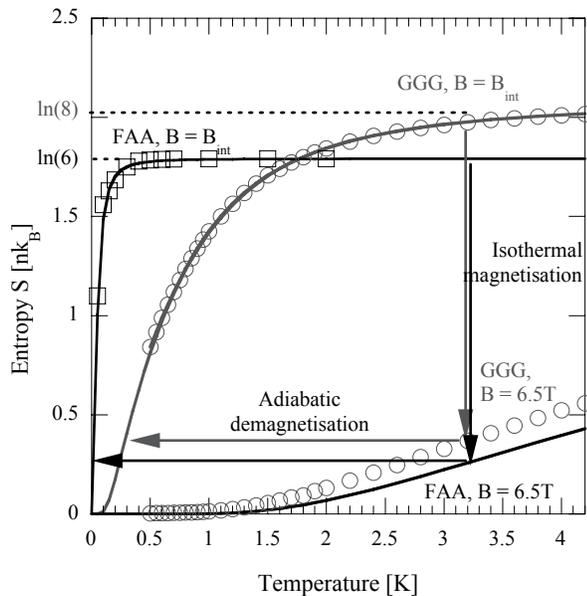


Fig. 5. Low-temperature entropy of ferric ammonium alum (FAA, black squares) and gallium gadolinium garnet (GGG, grey circles). The solid lines are theoretical values according to equation (2) with material constants from table (1). The arrows show the evolution of the entropy during an ADR cycle.

C. Detection Efficiency

The cold finger of the cryostat was re-designed to operate large detector arrays optimized for the energy range from 5 to 100 keV, with possible extension to ~ 200 keV or above [12]. The 0.1 K detector cold plate has an area of $4'' \times 4''$, larger than the ~ 5 cm² 112-pixel detector arrays that are currently operated in the cryostat to accommodate later spectrometer upgrades. The detector is held at 0.1 K behind four thin windows, one at 300 K, 50 K, 3 K and 0.1 K, respectively. The distance between the outermost window at 300 K and the detector is only 1 cm to ensure good solid angle coverage despite the small pixel size. The outermost window consists of 500 μ m beryllium, and sets the detection efficiency at lower energies (figure 6). It is designed for a transmission above 50% at 5 keV to allow measuring the actinide L X-rays in the 10 to 20 keV range. The spectrometer quantum efficiency for higher energy gamma rays is set by the thickness of the tin absorbers. Figure 6 shows the quantum efficiency for a 500 μ m Sn absorber, which is about $\sim 40\%$ in the 100 keV region that is important for nuclear analysis. For higher efficiency, future tantalum absorbers will provide an additional factor of ~ 2 improvement.

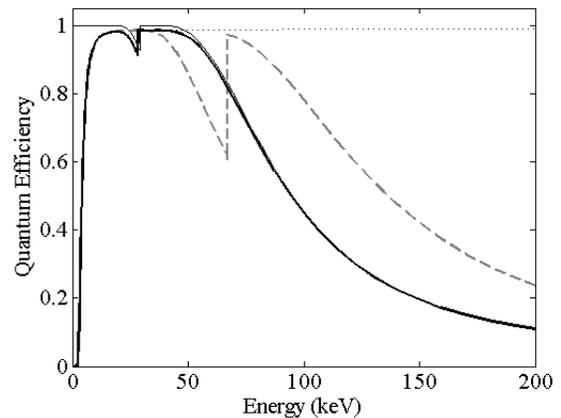


Fig. 6. The quantum efficiency of the TES spectrometer (thick black line) is set by the Be window transmission (dotted line) at low energies, and by the absorption of its Sn absorbers at high energy (thin lines). Future Ta absorbers will provide a factor ~ 2 improvement in quantum efficiency at ~ 100 keV.

D. Cryostat performance

The entire cryostat cool down is automated, and takes ~ 15 - 18 hours from room temperature due to the large mass of the ADR magnet and its magnetic shielding (figure 7). The ADR cycle uses a magnetizing current of 36 A for a maximum magnetization field of 6.5 T, and can be programmed ahead of time when the pulse tube has reached a temperature below 4 K.

Without regulation, the ADR attains a base temperature of ~ 28 mK, in agreement with the ordering temperature of 26 mK for the FAA paramagnet according to table (1). The GGG guard stage equilibrates around 380 mK, although this number depends somewhat on the initial temperature of the demagnetization cycle, as expected from figure 5. The cryostat base temperature is much lower than the ~ 0.1 K required for TES operation. We therefore use a small current in the ADR magnet to regulate the detector stage to the desired temperature. For an operating temperature of 0.1 K, the hold time between ~ 1 hour demagnetization cycles is greater than 48 hours with 32 signal wires connected to the TES detector stage.

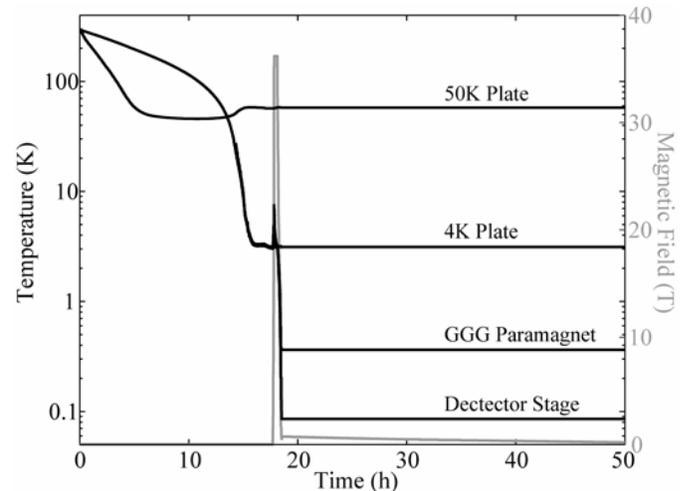


Fig. 7. Cooldown cycle of the pulse-tube ADR for regulation at 0.1 K

IV. SUMMARY

We have built a cryostat for superconducting γ -ray TES microcalorimeters that attains the required operating temperature below 100 mK without the use of liquid cryogens. It uses a two-stage pulse-tube refrigerator for precooling to ~ 3 K, and a two-stage adiabatic demagnetization refrigerator (ADR) to attain a base temperature of 28 mK. It is currently equipped with 32 signal wires connected to the detector stage to allow the readout of a multiplexed 112 channel TES γ -detector array. In this configuration, the cryostat has a hold time of more than two days between 1-hour demagnetization cycles for temperature regulation at a typical TES operating temperature of 100 mK. This instrument is a significant step towards making ultra-high energy resolution γ -ray spectrometers available to the wider nuclear analysis community since it no longer requires any expertise in low temperature physics.

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