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Cryogenic Microcalorimeter System for Ultra-High Resolution Alpha-Particle Spectrometry

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Abstract. Microcalorimeters have been shown to yield unsurpassed energy resolution for alpha spectrometry, up to 1.06 keV FWHM at 5.3 MeV. These detectors use a superconducting transition-edge sensor (TES) to measure the temperature change in an absorber from energy deposited by an interacting alpha particle. Our system has four independent detectors mounted inside a liquid nitrogen/liquid helium cryostat. An adiabatic demagnetization refrigerator (ADR) cools the detector stage to its operating temperature of 80 mK. Temperature regulation with ~15 uK peak-to-peak variation is achieved by PID control of the ADR. The detectors are voltage-biased, and the current signal is amplified by a commercial SQUID readout system and digitized for further analysis. This paper will discuss design and operation of our microcalorimeter alpha spectrometer, and will show recent results.

Keywords: microcalorimeter, alpha particle, transition-edge sensor
PACS: Replace this text with PACS numbers; choose from this list: http://www.aip.org/pacs/index.html

INTRODUCTION

Cryogenic microcalorimeters have been shown to achieve unprecedented high resolution for x- and gamma-ray spectroscopy, as well as alpha-particle spectrometry. (CITE) For analysis of low-activity samples, such as those encountered in nuclear forensics and environmental sampling, it is often most useful to perform alpha particle spectrometry rather than gamma-ray spectroscopy, as detection efficiency of alpha particles is much higher. However, conventional silicon detectors have insufficient energy resolution to resolve closely-spaced energy peaks from commonly encountered mixtures of radioisotopes. Therefore, expensive and time-consuming chemical separation may be required for meaningful isotopic analysis.

With a demonstrated resolution of up to 1.06 keV FWHM for 5.3 MeV alpha particles (CITE), it is possible to perform isotopic analysis with microcalorimeter detectors of samples containing mixed radionuclides that would be impractical to measure with conventional silicon detectors. Using many commercially available components, we have developed a four-channel cryogenic microcalorimeter system for alpha particle spectrometry at LANL. A similar one-channel system exists at NIST that is primarily used for detector development.

SPECTROMETER DESIGN AND OPERATION

Detector and Source Assembly

Microcalorimeter detectors use a superconducting transition-edge sensor (TES) to measure the temperature change in an absorber due to energy deposited by an incident photon or alpha particle. Figure 1 shows the detector assembly, which consists of the TES chip and first stage superconducting quantum-interference device (SQUID) amplifier. The TES consists of a molybdenum-copper bilayer, with a transition temperature of approximately 120 mK. During operation, the bath temperature is regulated at 80 mK, which allows the TES to be voltage biased into its transition region. Voltage bias creates a condition of negative electrothermal feedback, which makes the

TES stable against thermal runaway (CITE). Incident alpha particles deposit their energy in the absorber in the form of heat; therefore, unlike semiconductor detectors, a microcalorimeter detector has essentially no dead layer to introduce energy straggle. The absorber consists of a 4mm x 4mm x 0.25mm piece of tin and is mechanically and thermally coupled to the TES by multiple SU8 epoxy posts and has heat capacity C. The TES is supported by a thin SiN membrane, which provides a small thermal conductance G to the bath that allows each interacting alpha particle of energy E to produce a heat pulse in the absorber of height E/C, which decays with a time constant equal to C/G. (CITE) The TES therefore produces a current pulse proportional to this heat pulse, which is recorded for analysis.

In order to make full use of the high resolution and minimal straggle of the detector, alpha source preparation is critical. An ideal source is a uniform monolayer or less of alpha emitting radionuclides. Thick sources emit alpha particles with significant straggle due to energy loss within the sources material. Electrodeposition of radionuclides has been used successfully to make sources for measurement. (CITE)

The alpha particle source to be measured is situated millimeters from the detector in order to maximize detection efficiency. It is installed on the top cap of an aluminum cylinder that surrounds the detector assembly and provides shielding to avoid trapped magnetic flux in superconducting components. This shield is at the same temperature as the detector, which reduces radiant heat load on the absorber.

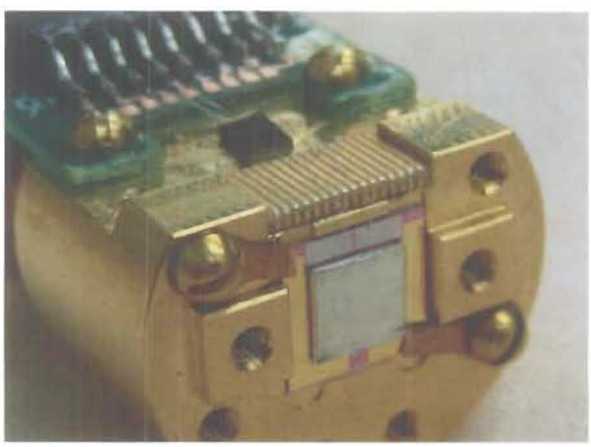

FIGURE 1. Detector assembly consists of TES chip with 4mm square Sn absorber and the first stage SQUID amplifier.

Readout Electronics

Each of the four detectors is read out through a separate channel, allowing four independent measurements to proceed simultaneously. As shown in figure 2, in each channel the bias current through the TES is inductively coupled to a superconducting quantum interference device (SQUID) amplifier chain that is manufactured by Star Cryoelectronics, Inc. (CITE). A first stage SQUID amplifier is mounted on the detector assembly to provide initial amplification. Further amplification occurs in a second stage SQUID series array, mounted on the 4K plate of the cryostat. Feedback circuitry, at room temperature, provides an error signal output that is digitized to provide a record of pulse shape for off-line analysis. All components of the readout electronics are commercially available.

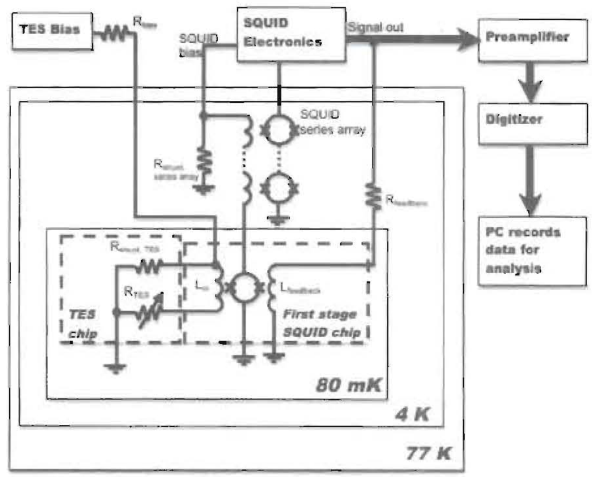

FIGURE 2. Block diagram of electronics for one channel

Data Analysis

Because the shape of each pulse is digitized and stored, much more information is available for analysis than if only individual peak heights are stored, as with typical commercial Si detector systems. This extra information can be used to decrease the effects of electronic noise and pulse pileup on energy resolution.

The energy resolution of a silicon detector is limited by the Fano statistics of electron-hole pair generation, but in a cryogenic microcalorimeter, thermal noise processes limit intrinsic resolution. With potentially over an order of magnitude better intrinsic resolution, electronic noise in the readout circuit becomes a significant factor in the ultimate energy resolution of a microcalorimeter detector system. Applying a Weiner-type digital filtering algorithm to the data, which applies measurements of system noise, can improve the energy resolution of the spectrometer.

Pulse pileup creates tailing on energy peaks in the measured spectrum, and decreases energy resolution. Because the shape of each pulse is digitized and stored for analysis, it is possible to separate piled up pulses with pulse fitting algorithms. These algorithms are in development, and will improve the ability of the system to measure higher activity samples.

Cryostat

The detectors are operated inside a liquid nitrogen- and liquid helium-cooled ADR (adiabatic demagnetization refrigerator) cryostat that is manufactured by Janis Research Company, Inc. (CITE). The ADR provides a maximum hold time of 20 hours at 80 mK. An AC resistance bridge and an analog PID controller manufactured by Stanford Research Systems, Inc. monitor the ruthenium-oxide resistance thermometer on the detector stage and adjust the magnet current to maintain a steady operating temperature (CITE). This control system provides better than 20 uK peak-to-peak temperature variation at the operating temperature of 80 mK. An external mu-metal can that surrounds the experimental space of the cryostat provides magnetic shielding that is required to reduce trapped magnetic flux in superconducting components. This trapped flux can change detector and SQUID amplifier response and must be minimized for stable operation.

RESULTS

209/210Po Spectra

209/210Po are useful isotopes for testing detector and source preparation performance. 210Po emits a 5304 keV alpha particle with nearly 100% intensity. This peak is especially useful to determine detector resolution and source straggle. Electrodeposited sources were made that contain 0.17 nCi of 209Po and 0.36 nCi of 210Po (activity on February 2, 2009). Source straggle is measured to be less than 2 keV. 209Po emits two alpha particles with similar energies: 4883 keV (80%) and 4885 keV (20%). As seen in figures 3 and 4, three different detectors with three different 209/210Po sources produce nearly identical spectra. The three alpha detectors installed for the measurement yield consistently better than 1.5 keV FWHM at 5304 keV.

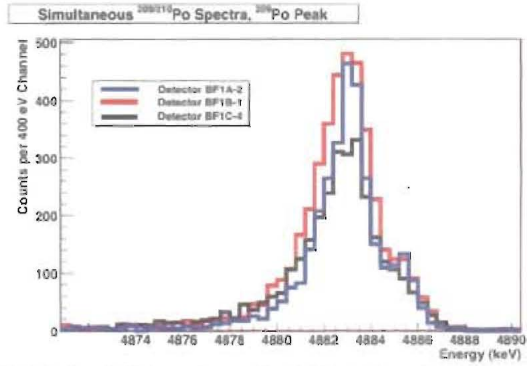

FIGURE 3. 209Po peaks, with 4885 keV peak visible as a shoulder on the more intense 4883 keV peak

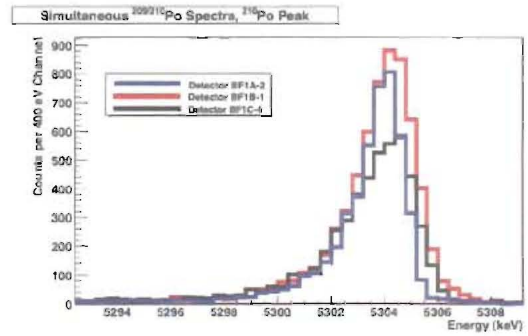

FIGURE 4. 210Po peak at 5304 keV

Mixed Actinide Spectrum

Isotopic analysis of nuclear material containing a mixture of actinides is a common challenge in nuclear forensics and environmental sampling. To examine the performance of a microcalorimeter detector in analyzing a sample with energy peaks too close to resolve well with a silicon detector, a sample containing 0.3989 nCi of 238Pu and 0.3426 nCi of 241Am (activity on October 20, 2008) was prepared. As seen in figure 5, the microcalorimetry mesurements provide sufficient resolution to determine peak height ratios even by eye, reducing the need for the complex fitting algorithms required for analysis of spectra from Si detectors and their associated errors. Preliminary results (March 2009) from the LANL spectrometer show comparable resolution to the spectrum from an earlier measurement (May 2008) with the NIST system. This demonstrates that the detector technology is robust enough to provide consistent results with completely different cryostats and readout electronics.

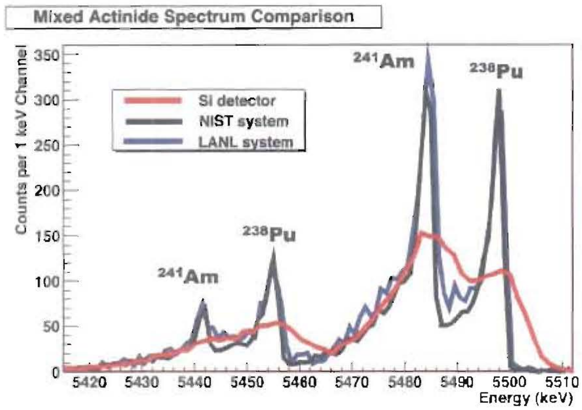

FIGURE 5. Spectrum of sample containing both 241Am and 238Pu, as measured on the NIST and LANL microcalorimeter systems

Instrument Stability

Over a 20 hour measurement, as limited by the ADR hold time, there is no significant variation in detector response or loss of SQUID amplifier lock as seen in figure 6. Some variability in detector response occurs between ADR magnet cycles due to irregular flux trapping. The aluminum shielding (Tc=1.1K) around each detector assembly undergoes a superconducting transition on each ADR magnet cycle while there is some stray field from the magnet and therefore can produce variable flux trapping. Niobium shielding (Tc=9.2K) will become superconducting when the cryostat is initially cooled and there is no field from the ADR magnet. New shields made of niobium are currently being tested.

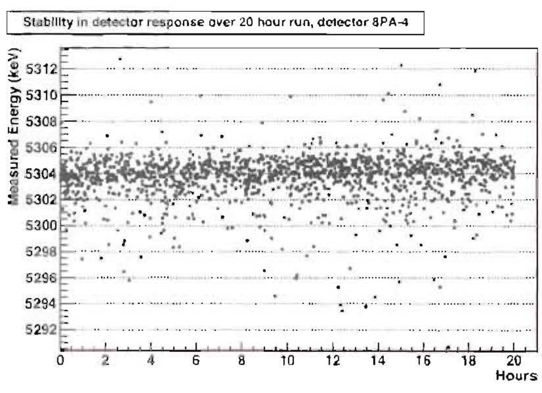

FIGURE 6. Measurement of 210Po shows no significant variation in detector response or loss of SQUID amplifier lock over a period of 20 hours

FUTURE WORK

The current four-channel spectrometer will be used for measurement campaigns to evaluate the performance of alpha source preparation techniques and new detector designs for alpha particle and gamma-ray spectroscopy. Detectors with improved counting efficiency and faster response will decrease required measurement time.

The wet cryostat will be upgraded to a mechanically cooled ADR system like the LANL gamma spectrometer, which uses no liquid cryogens. In addition to yielding more reliable performance and simpler operation, the dry cryostat will improve sample throughput due to its shorter cooling time. Eight independent channels are planned for this system.

At present, installation of alpha sources requires that the cryostat be warmed to room temperature and partially disassembled. This is a complicated and time-consuming procedure. To solve this problem, a cryogenic load lock is currently in development that will allow samples to be installed while most of the cryostat remains cold and under vacuum. Multiple samples will be installed in a holder that can be gradually lowered into position next to the detectors. As the sample holder is lowered, heat switches will be connected in sequence to cool it to 60K and finally 3K when it is positioned adjacent to the detectors.

FIGURE 7. Proposed design for cryostat with load lock for rapid sample exchange

ACKNOWLEDGMENTS

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