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Ultra-high resolution alpha particle spectrometry with transition-edge sensor microcalorimeters

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Alpha-particle spectrometry is a powerful analytical tool for nuclear forensics and environmental monitoring. Microcalorimeter detectors have been shown to yield nearly an order of magnitude better energy resolution (1.06 keV FWHM at 5.3 MeV) than current state-of-the-art silicon detectors (8-10 keV FWHM at 5.3 MeV). This superior resolution allows isotopic analysis with a single non-consumptive measurement of samples that contain multiple radioisotopes with overlapping alpha energies. Measurement of such a sample with a silicon detector would require expensive and time-consuming radiochemical separations. We are developing two alpha spectrometer systems with superconducting transition-edge sensor microcalorimeters. Each uses a helium pulse tube cryostat with an adiabatic demagnetization refrigerator to allow operation of the detectors at ~100 mK. The first system has eight independent detector channels that measure eight different alpha sources, and is optimized for detector development experiments. The second system incorporates a prototype cryogenic load lock that allows for rapid exchange of alpha sources. This paper will present results from these two systems.

1. INTRODUCTION

Nuclear forensics and environmental monitoring require rapid isotopic analysis of trace actinide-containing samples, for which alpha-particle spectrometry is a particularly powerful analytical tool. Microcalorimeter alpha-particle detectors have been shown to yield nearly an order of magnitude better energy resolution (1.06 keV FWHM at 5.3 MeV) than current state-of-the-art

silicon detectors (8-10 keV FWHM at 5.3 MeV)[1]. This superior resolution allows rapid isotopic analysis with a single non-consumptive measurement of samples that contain multiple radioisotopes with overlapping alpha energies. Measurement of such a sample with a silicon detector would require expensive and time-consuming radiochemical separations. We are developing two microcalorimeter alpha spectrometer systems in order to bring this technology towards the goal of an analytical instrument that can be used for rapid isotopic analysis of these trace samples.

2. MICROCALORIMETER SYSTEM FOR DETECTOR, SOURCE AND DATA ANALYSIS DEVELOPMENT

From late 2008 to early 2011, we developed and operated a four-channel microcalorimeter alpha spectrometer at LANL for the development of improved microcalorimeter detectors, high-resolution alpha source preparation methods, and data analysis techniques[2,3]. This system has now been replaced with an improved eight-channel version that is similar in concept, but has expanded capabilities and performance. Primary design goals for the eight-channel system are reliable and stable cryogenic operation, and a flexible internal configuration to facilitate experiments for the development of improved detectors, sources, and data analysis.

A key feature of the LANL four-channel system was its modular design. Interchangeable mounts enabled convenient assembly and testing of new detectors and sources. This same design is the basis for the eight-channel system. The eight-channel system is built around a High Precision Devices model 106 helium pulse-tube cryostat with an adiabatic demagnetization refrigerator (ADR).[4] Unlike the previous system, it uses no liquid cryogens and is therefore simpler to operate. The Ø 34cm by 21cm tall 3K experimental space provides sufficient volume for eight independent detectors and alpha sources, with extra space to accommodate future modifications. Cooling the detector stage from room temperature to 3 K takes less than 15 hours. Magnetic shielding, integrated into the cryostat vacuum jacket, reduces the ambient magnetic field at the detectors by more than a factor of 50. A superconducting aluminum can that surrounds each detector assembly provides further magnetic shielding. The ADR is operated by a semi-automated computer control system that incorporates error monitoring, and simplifies operation.[5] We have demonstrated continuous detector operation at 80 mK with less than 10 microkelvin variation for greater than 50 hours, limited by the ADR.

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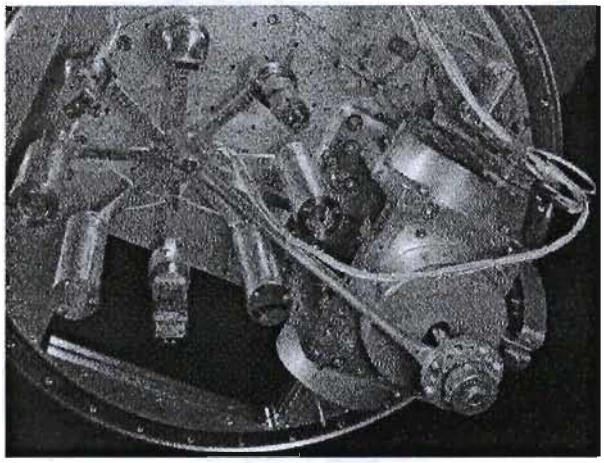

Fig. 1. Eight channel detector stage, with modular detector and source mounts in various stages of disassembly.

Fully independent detectors, sources, and readout allow for experiments on individual channels. Each detector is read out with a STAR Cryoelectronics two-stage SQUID amplifier.[5] Voltage bias for the detectors, data acquisition, and instrument control functions will be provided by a National Instruments PXI system.[6] By integrating much of the instrument control electronics with this PXI system, we will have great flexibility in testing. The PXI system incorporates a 24-bit ADC that should simplify data acquisition throughout the dynamic range of the detector, which has been demonstrated to provide spectroscopic resolution for ~10 keV x-rays and ~5000 keV alpha particles in a single measurement.

This eight-channel system has demonstrated alpha energy resolution comparable to the best that we have achieved. A 24-hour measurement of a ^{210}Po source yielded a resolution of 1.23 keV FWHM at 5.3 MeV. The detectors used for this measurement are of a design that has been used for several years in the previous four-channel spectrometer. They consist of a Mo/Cu bilayer TES with a transition temperature of approximately 120 mK, and a 4 mm square by 0.25 mm thick bulk Sn absorber attached by epoxy posts.[2]

In order to make alpha detectors for this system, a batch of TESs were fabricated at NIST. These devices are identical to previously made (type A) TESs, with the exception of the SiN membrane thickness. By using 1.5 micron

rather than 0.5 micron SiN as in the previous design, the new devices are substantially more robust. We have demonstrated that the 1.5 micron membrane can support a pressure difference of at least 78 kPa (11.3 psi). However, the thermal conductance of the membrane G is now a factor of three higher. The result is that the current through the TES must be higher to reach the same bias point in the transition.

The devices with 1.5 micron membranes (type B) were found to be unstable at many bias points throughout their transition. The instabilities, shown in figure 2, are characterized by a rapid switching between two levels and depend strongly on the bias point of the TES. Their structure is very different from that of electrothermal oscillation, where the TES response with electrothermal feedback is underdamped.[7] At higher bias currents within the superconducting transition, corresponding to bias points at a lower fraction of the TES's normal state resistance R_N, the instabilities are more pronounced. Some of these devices are unstable at bias points from 0 to 87% R_N. With such a large fraction of the superconducting transition corresponding to an unstable detector response, the usable dynamic range of these devices is insufficient for high-resolution alpha spectrometry.

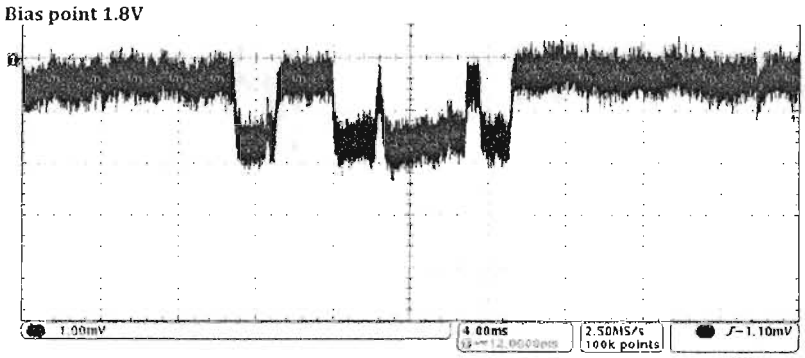

Fig. 2. Instability in detector response produces spontaneous switching between levels when biased at certain points in the superconducting transition.

Testing of previous devices with a 0.5 micron membrane revealed similar instabilities at bias points from 0-20% R_N. Although this portion of the transition must be avoided for stable detector operation, there is sufficient usable dynamic range available in these devices for successful alpha spectrometry.

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Comprehensive testing of a number of old and new devices demonstrated that, other factors constant, the instabilities are most pronounced for devices with higher transition temperature T_C and higher G. For a given device, the instabilities are most pronounced when biased at a lower fraction of R_N or when operated at a lower bath temperature. These characteristics lead us to the hypothesis that the underlying cause for the instabilities is an excessive current density in the TES. The switching behavior may arise from regions of the device oscillating between normal and superconducting as the bias current switches paths.

To test this hypothesis and attempt to create more stable devices, a new batch of TESs on 1.5 micron membranes were fabricated. Along some type B devices as a control, a third variety (type C) was fabricated with the TES bilayer scaled to reduce the bias current density by a factor of three (figure 3). Preliminary tests indicate that while these type B devices are unstable at bias points up to 53% R_N, the type C devices only show instabilities below 20% R_N. These results indicate that the type C devices can provide the robustness of a 1.5 micron SiN membrane with stability similar to type A devices. Further testing is ongoing to determine the alpha spectrometry performance of these type C devices.

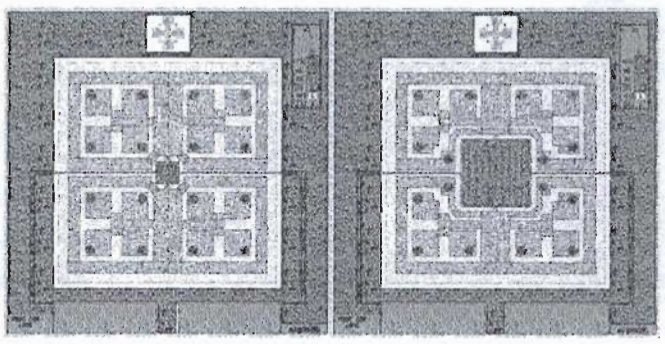

Fig. 3. Original alpha TES design on left, used for type A and B devices, compared with type C device on right. Type C has an enlarged Mo/Cu bilayer (center square) to reduce bias current density by a factor of 3.

3. MICROCALORIMETER SYSTEM WITH CRYOGENIC LOAD LOCK

Alpha spectrometry with cryogenic detectors presents a unique challenge due to the non-penetrating nature of alpha particles. The alpha source and detector must be mounted together in vacuum, and the source held at a low enough temperature to avoid excessive radiative heat load on the detector. In the eight-channel system, sources are attached directly to the detector stage. While this simple mounting scheme provides good geometric efficiency and thermal and magnetic shielding for the detector, it requires that exchange of sources is done with the entire cryostat warm and open. Due to the time required to warm and cool the entire cryostat, it is not practical to measure more than two batches of sources per week in this system.

An important requirement of a practical instrument for alpha spectrometry is simple, rapid sample exchange. To achieve this goal, we are developing a cryogenic load lock in a second microcalorimeter alpha spectrometer system. By keeping most of the cryostat cold, source insertion time is reduced to hours instead of days.

The prototype cryogenic load lock is currently being developed at NIST in a dry ADR cryostat identical to the one used for the LANL 8-channel spectrometer. With the cryostat cold, up to four alpha sources at a time are attached to a source holder at the end of an insertion rod. A standard vacuum load lock allows the sources to enter the cryostat vacuum space. The source holder is lowered into the cryostat, and clamped to the 50 K stage to begin cooling. After this initial cooling, the 50 K clamp is released and the source holder is lowered and clamped to the 3 K stage. With the source holder in the measurement position, clamped at 3 K, the insertion rod can be pulled back above the 50 K stage, and 3 K and 50 K shutters closed to prevent excess heat load on the detector stage. Figure 4 shows some key components of the load lock.

With the sources held at 3 K, the ADR can be cycled to cool the detector stage to its operating temperature of approximately 80 mK, and the detectors operated as in the 8-channel spectrometer system. Early results show that the source holder can be cooled to 5K in approximately 60 minutes. This decreased source insertion time allows the measurement of one set of sources per day, rather than two sets per week as with the 8-channel development spectrometer. In the LANL four-channel spectrometer system, we have demonstrated that thermal load from a 3 K source is not enough to significantly degrade detector performance. Testing of the cryogenic load-lock with a detector and source is in progress.

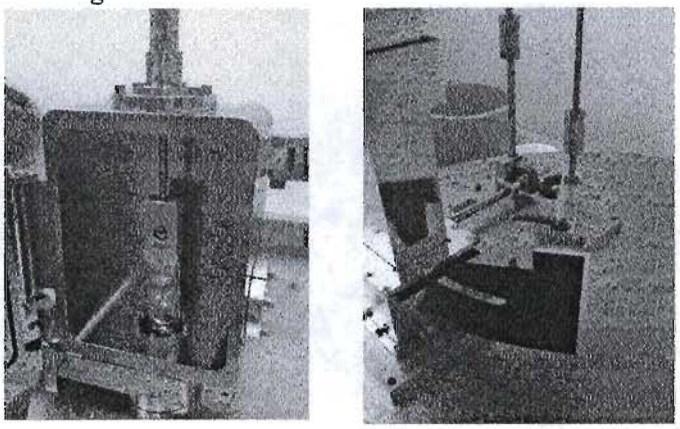

Fig. 4. Cryogenic load-lock components include vacuum load-lock with sample holder (left) and shutter/heat sink assembly (right).

4. FUTURE WORK

Testing of the type C TESs in the LANL 8-channel spectrometer system is ongoing. If this TES design performs well for high-resolution alpha spectrometry, it will be used as the basis for alpha detectors in this system. A PXI-based instrument control and data acquisition system is being implemented that will increase the capability and usability of the spectrometer. One of the major challenges of using microcalorimeter alpha spectrometry for isotopic analysis of actinide samples is insufficient understanding of the alpha energy peak shape. Peak-shape models developed for silicon alpha detectors do not describe microcalorimeter data well enough to avoid significant systematic bias when determining overlapping peak amplitudes. Improved detectors and data acquisition will facilitate measurement campaigns of complex alpha sources to help develop peak-shape models optimized for microcalorimeters.

The prototype cryogenic load lock mechanism is continuing to be improved for more efficient operation. Installation and testing of all four detector channels will enable measurement of up to four alpha sources simultaneously. Once the load-lock spectrometer has demonstrated the ability to generate high-resolution alpha spectra, it will be a significant step towards a practical analytical instrument for rapid isotopic analysis of trace actinide samples.

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