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Nuclear diagnostics with cryogenic spectrometers

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Abstract

Cryogenic spectrometers offer an energy resolution an order of magnitude higher than conventional high-purity Germanium detectors, since low-temperature operation reduces thermal fluctuations and the associated noise. They have been developed over the last several decades, initially driven by particle and astrophysics applications, more recently also for material science, biophysics and nuclear science applications. The Advanced Detector Group at LLNL is developing cryogenic Gamma-ray and fast-neutron spectrometers for nuclear diagnostics in fundamental science and national security. They are based on measuring the increase in temperature upon photon or particle absorption with a sensor operated at the transition between its superconducting and its normal state. The approach can be adapted for different types of radiation with the appropriate choice of absorber material. We describe our Gamma-ray spectrometers consisting of a bulk Sn absorber attached to a superconducting Mo/Cu multilayer sensor. The detectors are operated at a temperature of ~0.1 K the end of a cold finger in a two-stage adiabatic demagnetization refrigerator, and have, depending on design, achieved an energy resolution between 50 and 150 eV FWHM for energies below 100 keV. Here we give an overview about cryogenic detectors for nuclear diagnostics, and discuss the application of this detector technology to the measurement of uranium enrichment.
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1. Introduction

Calorimetric measurements have a long history in nuclear science, going back to Curie's seminal experiments that confirmed that heat released by a radioactive substance is due to the energy of the associated particle emission [1]. It was first pointed out by Simon in 1935 that the sensitivity of calorimeters for nuclear science can be improved by sensor operation at low temperature where heat capacities and thermal fluctuations are low [2]. This led to the development of superconducting detectors in the 1940s, both as bolometers for infrared radiation and as alpha-particle detectors, with the difference of the two detectors being due to different absorbers optimized for the radiation of interest [3,4]. Later, semiconducting Si and Ge detectors were developed as alternative cryogenic sensors, with the choice of sensor material often determined less by its intrinsic merit but by its manufacturability and ease of operation [5].

Cryogenic detector development has increased since the 1980s, when the required technology investment was motivated by high-profile scientific questions, such as neutrino mass physics [6,7], dark matter searches [8], X-ray astronomy [9] and material analysis [10]. This led not only to the development of different detector types with complementary characteristics with respect to energy resolution, active volume and maximum count rate, but also to mature readout and refrigeration technology required to operate these detectors at temperatures below 1 K [11].

At Lawrence Livermore National Laboratory, we have been developing cryogenic calorimeters for nuclear diagnostics and high-energy astrophysics for two decades. Initially, our focus was on neutron transmutation-doped (NTD) Ge sensors [12]. Later, we have focused on superconducting detectors because their photolithographic device fabrication allows comparably easy scaling to large

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arrays [13–16]. Here we discuss the performance of our superconducting Gamma (γ) detectors, their use in nuclear diagnostics to measure uranium enrichment, and the trade-offs involved in optimizing detector sensitivity.

2. Experiment

For ultra-high resolution γ-spectroscopy, the Advanced Detector Group at LLNL is developing superconducting Mo/Cu multilayer transition edge sensors (TESs) with attached bulk absorbers [14,15]. Currently, the absorber material of choice is superconducting tin despite its comparably low atomic number $Z = 50$ and thus only moderately high absorption efficiency, because it currently still offers the best energy resolution of ~50 eV FWHM [16]. The 99.999% pure tin foil absorber is strongly coupled with epoxy to a small $500\,\mu m \times 500\,\mu m \times 0.2\,\mu m$ Mo/Cu multilayer TES, which in turn is weakly coupled to the cryostat cold stage though a thin free-standing silicon nitride membrane. To first approximation, the energy resolution of such a detector is determined by the heat capacity C_{abs} of the bulk tin absorber according to [9]

$$\Delta E_{FWHM} \approx 2.355(k_B T^2 C_{abs})^{1/2}, \quad (1)$$

where k_B is the Boltzmann constant and T is the absolute temperature. An operating temperature of $T \approx 0.1$ K provides a good compromise between high energy resolution and ease of operation, since it can be attained comparably easily with the use of adiabatic demagnetization refrigerators (ADRs) [17]. We have built several spectrometers for different cryogenic detector technologies that use liquid N_2 and He to precool to 77 and 4.2 K, respectively, and a two stage ADR to attain a base temperature of ~70 mK with a hold time up to ~24 h per demagnetization cycle, depending on the heat load and thus the number of wires to the cold stage [15,17]. In these spectrometers, the detector is held at the end of a shielded cold finger adapted for the particular scientific application, so that the intrinsically small detectors can be operated at ~0.1 K within ~10 mm of a sample at room temperature. Since the impedance of TES detectors is below 1 Ω, the signal readout requires a superconducting quantum interference device (SQUID) preamplifier, operated in a magnetic shield inside the cryostat and read out by a room temperature main amplifier in an electromagnetically shielded enclosure attached directly to the cryostat (Fig. 1).

As an illustrative example for potential of cryogenic detectors for nuclear diagnostics, for the potential we have exposed a cryogenic γ-detector to a weak source of low-enriched uranium (LEU) for ~48 h. The interest here is to determine the level of ^{235}U enrichment above the natural concentration of 0.7202%. The ^{235}U/^{238}U isotope ratio can be extracted from the measured line intensity ratio of the ^{234}Th γ-emission at 92.38 and 92.79 keV, and the Th $K_{\alpha 1}$ X-ray line at 93.35 keV [19,20]. The Th $K_{\alpha 1}$ X-rays are mostly due to the decay of ^{235}U [18,19], and the ^{234}Th emission provides a measure of the ^{238}U concentration, provided the uranium sample is at least ~200 days old and thus in secular equilibrium with its thorium daughters.

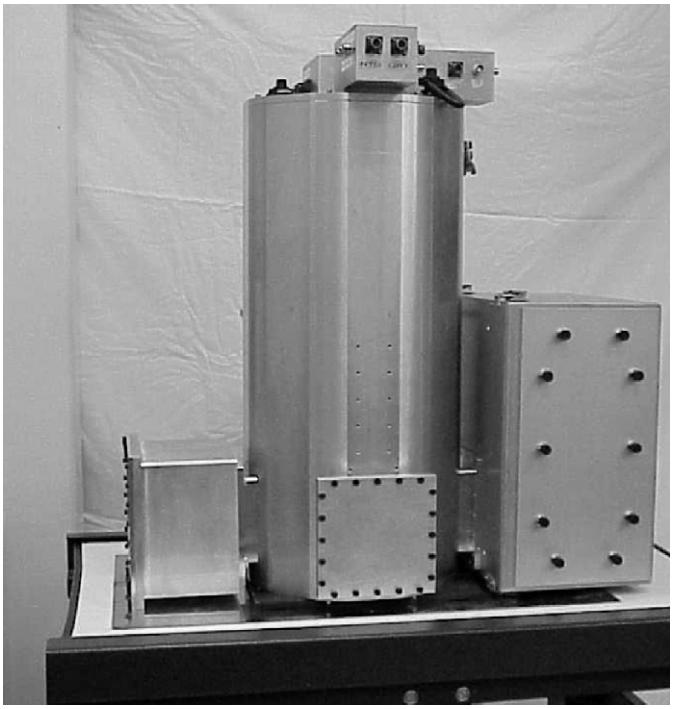

Fig. 1. Spectrometer developed for operating superconducting detectors at ~0.1 K the end of a cold finger (left) within ~10 mm of a room T sample.

The advantage of measuring uranium enrichment using these emission lines lies in their similar energies, which reduces systematic errors from variations in detection efficiency at different energies due to self-absorption, shielding transmission and detector quantum efficiency. On the other hand, the separation of these lines by only 551 eV places stringent requirements on the energy resolution of the spectrometer to reduce the errors due to line overlap, especially when the lines have very different intensities.

Cryogenic detectors do have sufficient energy resolution to resolve the ^{234}Th γ-rays and the Th $K_{\alpha 1}$ X-ray above the Compton background (Fig. 2). In fact, since the energy resolution of 90 eV FWHM for this detector allows a full separation of the two ^{234}Th γ-rays, these measurements can be used to reduce the error bars on the associated branching ratios. Note that the X-ray lines in this spectrum clearly appear broadened compared to the γ-rays because of the life time broadening of ~90 eV for the associated core holes in Th and U. For comparison, we have included a γ-spectrum of the same sample taken with a planar high-purity germanium (HPGe) detector over ~24 h. While the energy resolution of ~600 eV FWHM is much lower than that of the cryogenic TES detector, its quantum efficiency is much higher due to the larger pixel size. This improves the statistics of the measurement and allows applying peak stripping techniques to extract isotope ratios from γ-spectra despite line overlap. For this sample, assuming