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Magnetic Microcalorimeter (MMC) Gamma Detectors with Ultra-High Energy Resolution

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Magnetic Microcalorimeter (MMC) Gamma Detectors with Ultra-High Energy Resolution

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Project Goal

The goal of this LCP is to develop ultra-high resolution gamma detectors based on magnetic microcalorimeters (MMCs) for accurate non-destructive analysis (NDA) of nuclear materials. For highest energy resolution, we will introduce erbium-doped silver (Ag:Er) as a novel sensor material, and implement several geometry and design changes to improve the signal-to-noise ratio. The detector sensitivity will be increased by developing arrays of 32 Ag:Er pixels read out by 16 SQUID preamplifiers, and by developing a cryogenic Compton veto to reduce the spectral background. Since best MMC performance requires detector operation at ~10 mK, we will purchase a dilution refrigerator with a base temperature <10 mK and adapt it for MMC operation. The detector performance will be tested with radioactive sources of interest to the safeguards community.

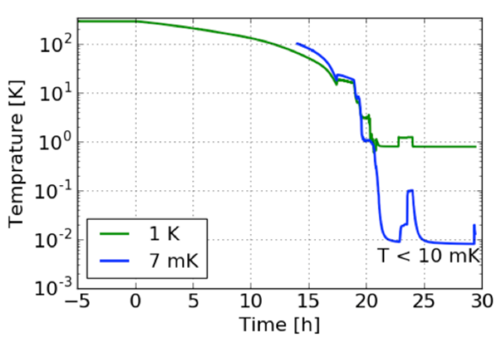
In FY16, we specced and purchased a liquid-cryogen-free dilution refrigerator for MMC gamma detector operation, and finalized the process development to fabricate Ag:Er-based MMCs. We also built two-pixel MMC prototypes and tested them with different radioactive sources.

The goals for FY17 were then 1) to modify the dilution refrigerator for operation of MMC arrays, 2) to fabricate the first MMC detector arrays based on the results of FY16, and 3) to test these arrays with radioactive sources of interest for nuclear safeguards and demonstrate an energy resolution below 100 eV FWHM. This report discusses progress in FY17.

1) Dilution Refrigerator Modifications for MMC

The primary specifications for the dilution refrigerator were a base temperature below 10 mK (below 8 mK if possible), and a cooling power above 10 µW at 20 mK (ideally above 15 µW at 20 mK). User-friendliness and low-cost operation implied that the cryostat should attain these values without the use of liquid nitrogen or liquid helium, which can be achieved by using a pulse-tube refrigerator for precooling to 3 K. Since the associated compressor and rotary valves emit significant electromagnetic signals, the cryostat should be electromagnetically shielded and galvanically isolated form the pulse tube. Similarly, vibrations from the pulse tube should be isolated as much as possible from the cryostat cold stage to minimize microphonic noise. Finally, the cryostat needed to have a side port to allow placing a radioactive source close to the (intrinsically small) MMC detector, and it should work reliably with a minimum of maintenance.

The BF400 dilution refrigerator (Figure 1) that we purchased from Bluefors Cryogenics, a small company in Finland, fulfils these challenging requirements beautifully. It has reached a based temperature of ~7 mK, well below the required specification (Figure 2), and its cooling



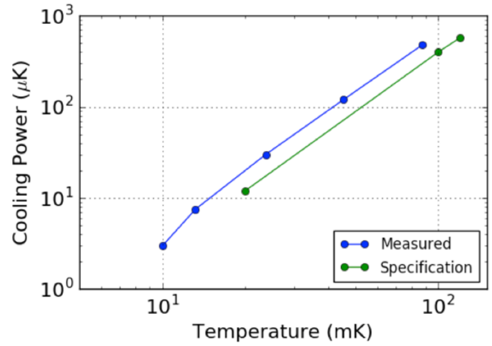


Fig. 1 (left): Dilution refrigerator with side port behind which MMC detector is operated. Fig. 2 (center): Automated cooldown of the dilution refrigerator to its base temperature <10 mK within 24 hours. The green line is the thermometer signal of the still, the blue line is the temperature on the MMC detector stage. Fig 3. (right): The measured cooling power (blue) exceeds the specified cooling power (green) significantly.

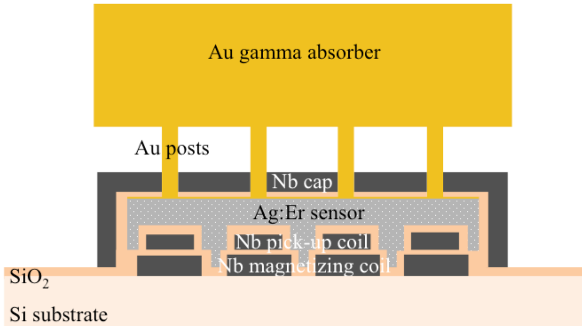
power of 20 µW at 20 mK exceeds the requirement by a factor of 2 (!) (Figure 3). The cooldown is automated and does not require the use of cryogenic liquids, and the associated electromagnetic interference and mechanical vibrations by the pulse tube refrigerator –while noticeable– do not preclude low noise measurements.

We have then upgraded the wiring and the preamplifiers inside the dilution refrigerator to accommodate the readout for 32 detector pixels. This included the installation of 16 SQUID arrays preamplifiers at 4 K to read out signals from 32 MMCs, 16 channels of feedback electronics on the cryostat frame to bias the SQUIDS, and several multi-wire “looms” with heat sinks for the electrical connections. We have also modified the windows of the side port to place the radioactive source close the MMC detector array. Currently, the outermost window suffers from a leak in the weld joint, which will be fixed in FY18.

2) Fabrication of Ag:Er MMC Gamma Detector Arrays

Most of the photolithographic process development to fabricate MMC gamma detectors had occurred in FY16 and has been summarized in the FY16 reports. This development included fabricating the SQUID preamplifiers on a 4” Si wafer using a modified version of STAR Cryo’s Delta-1000 process, dicing the wafer into 1 cm^2 chips for deposition of the paramagnetic Ag:Er sensor, and electroplating the thick Au absorber onto these sensors.

The one step that turned out to be much more difficult than expected was the fabrication of a gold absorber on top of the paramagnetic Ag:Er sensor. The difficulty arises from the fact that the absorber needs to be supported on posts to ensure that the gamma energy thermalizes uniformly in the absorber before it heats the Ag:Er sensor (Figure 4). This requires the deposition and patterning of two different photoresists (one for the posts, one for the absorber) whose chemistries cannot interfere with one another. Despite significant effort, we have not been able to reliably implement the two-stage process with AZ 125 nXT photoresist that our graduate student Cameron Bates had developed in Heidelberg as part of the predecessor proposal. The attempt to combine the AZ 125 nXT for the absorber with a sacrificial copper layer for the posts also failed, because film stress in the copper would damage the underlying structures. We eventually came



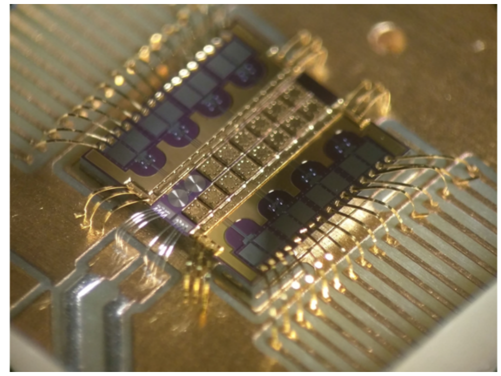


Fig. 4 (left): Schematic cross section of the MMC with the Au gamma-ray absorber supported on top of the Ag:Er sensor by Au posts. Current in the lower Nb coil magnetizes the Er spins in the Ag:Er sensor, and sensor heating by gamma-rays induces a signal in the upper Nb pick-up coil. Fig. 5 (right): Picture of the 2×8-pixel MMC array with 8 SQUID preamplifiers mounted on an alumina chip carrier. The two leftmost pixels do not have an absorber on top of the round pick-up coil because they are reconfigured to serve as an on-chip thermometer. The superconducting Al wirebonds on the left provide the current for the Nb magnetizing coil, and the Au wirebonds next to them serve as thermal anchors to ensure fast thermal decay of the MMC signal.

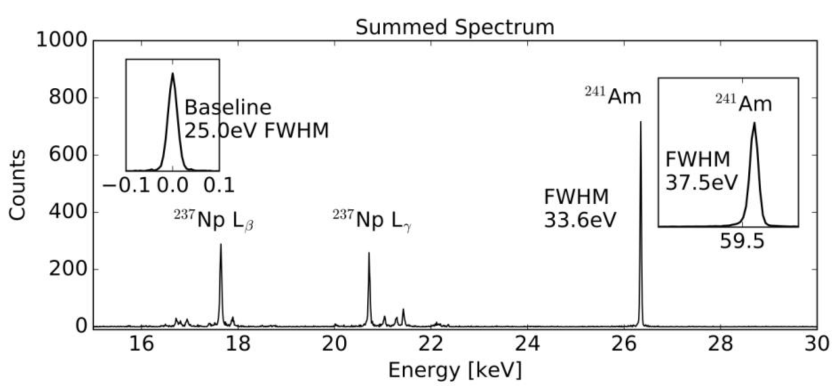
up with a process to fabricate the Au absorber using the MX5000 series of dry-film photoresists from DuPont. We used a 20 µm thick MX5020 resist for the posts, and a separately patterned MX5050 resist for the thicker Au absorber on top. The resist is rolled onto sample on a hot plate at 90-100 ºC, with gentle pressure applied by a custom hand roller. It is then exposed to 365 nm UV light at an intensity of 18 mW/cm^2, and developed in a K_2CO_3 solution at 30 ºC. The Au posts and absorbers are electroplated in a single step from a fresh Technic D7990 RTU solution with a Au density of 8.2 g/l under agitation with water pump directed toward the chip. After plating, both photoresist layers are removed by ultrasonic agitation in dimethyl sulfoxide (DMSO) at 50 – 60 ºC. Details of the process are described in the paper by graduate student Ruslan Hummatov who led the fabrication effort. Figure 5 shows a picture of the completed MMC array mounted onto a chip holder. The 14 squares in the center of the chip are the Au absorbers, and the posts underneath are visible as a ring of small dots. The absorbers have a uniform thickness across the cm-scale chip, a residual resistance ratio of ~7, and are quite robust, surviving multiple rapid immersions into liquid nitrogen and long ultrasonic activation in different solvents. The signals from our initial MMC gamma detector fabricated with this process have rise time of ~ 7 µs and show no signs of spatial variation across an absorber. This suggests that the absorber thermalizes the gamma energy well.

But even thick electroplated Au absorbers do not offer much efficiency for gamma-rays above ~200 keV. This is acceptable, because ultra-high energy resolution is mostly needed for lower-energy gamma-rays where line overlap is strongest. But it would still be desirable to record higher-energy gamma-rays rather than losing the information they carry. We therefore intend to operate a high-purity germanium (HPGe) in the final instrument directly behind the MMC to capture transmitted gammas. This HPGe detector would not only extend the energy range of the instrument to >1 MeV, it would also serve as a Compton for gamma-rays that are forward-scattered in the MMC and thereby reduce the spectral background at low energy. The question is whether HPGe detectors can be operated at ~10 mK without loss in energy resolution.

In FY16, we observed that the energy resolution of our HPGe detector degraded over time at temperatures <0.5 K. This is because residual impurities in the detector freeze out and slowly form traps that can capture gamma-induced charges. We tried to address this in FY17 by mounting an LED next to the Ge detector in one of our older ADR refrigerators and periodically flash it with visible light to refill the traps. This procedure that has allowed the Cryogenic Dark Matter Search (CDMS) to operate their HPGe detector at 10 mK for months. Although the Ge detector could retain its energy resolution over more than a week, the experiments were ultimately inconclusive. This is because the Joule heating of the LED would warm up the Ge detector well above 10 K whenever we tried to activate the LED, and it was not clear whether the good energy resolution was due to the absorbed photons or due to heating the Ge close to 77 K. This type of heating would damage our dilution refrigerator. We will therefore install an optical fiber in our dilution refrigerator in FY18 so that we can expose the Ge detector to optical photons from an external source without heating it overly strongly. If this is not successful and its energy resolution still slowly degrades over time, the Ge detector should still be able to function as a Compton veto. This would satisfy the initial goal of this LCP, although it would be somewhat disappointing not to be able to see higher-energy photons.

3) Detector Performance

We have systematically tested the MMC detectors with radioactive sources in our dilution refrigerator at different temperatures and magnetizing currents. For low-noise operation we had to use Kapton tape and plastic screws to insulate the MMC galvanically from the rest of the cryostat, which serves a giant antenna and picks up all kinds of electromagnetic interference from the environment. As expected, the best performance was then obtained at the highest magnetization current of ~100 mA through the Nb magnetizing coil below the Ag:Er sensor. We have measured an energy resolution as high as 38 eV FWHM at 60 keV (Figure 6). This is a world record for MMC detectors at this energy, and already exceeds the FY18 performance goal of this LCP. Interestingly, the resolution can likely be further improved, because the MMC was not at the base temperature of the cryostat. Although the cryostat itself was at a temperature <10 mK, the power dissipation from the SQUID preamplifier would heat the MMC that was integrated on the same chip to a temperature of several ~10 mK and reduce its resolution. Future devices should therefore improve heat sinking or separate MMC and SQUID onto two chips linked by superconducting Al wirebonds. We will explore these trade-offs in FY18.



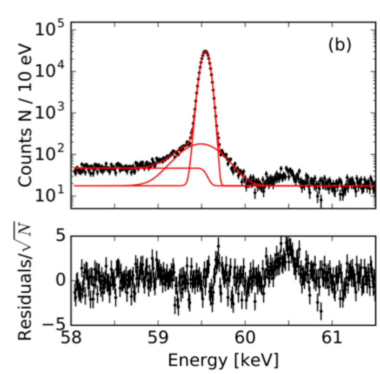
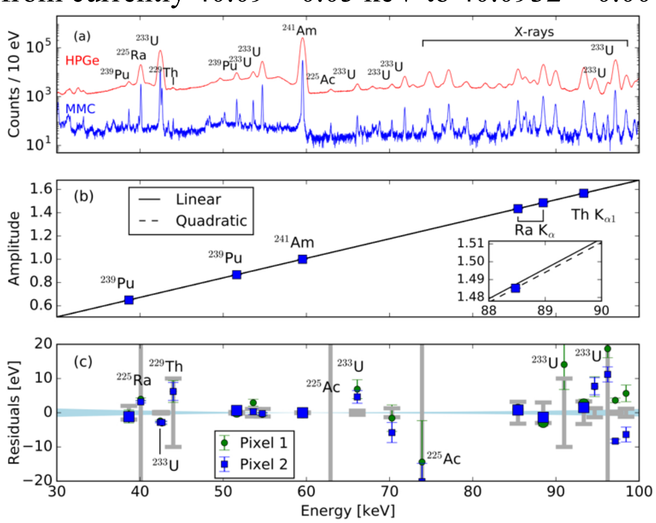


Fig. 6 (left): High-resolution spectrum of an Am-241 source taken with the MMC gamma-ray detector in figure 5.
Fig. 7 (right): The response function of the MMC gamma detectors from our collaborators at Heidelberg University has a Gaussian response on top of a step background due to small angle scattering between source and detectors as expected. In addition, there is a small distribution of events due to pile-up of dual-polarity pulses.

We also have access to MMC gamma detectors from our collaborators at Heidelberg University. These detectors have thicker Au absorbers and therefore higher detection efficiency, albeit at the expense of lower energy resolution, around 100 eV FWHM in this experiment. Figure 7 shows the response function of the Heidelberg MMCs at 60 keV. It consists of a primary Gaussian peak on top of a step background due to small angle scattering between source and detectors as expected. In addition, there is a small distribution of events that we can attribute to pile-up. It extends to both sides of the primary peak since the gradiometric design of our MMCs produces pulses with both polarities. We can fit the response function at 60 keV to a sum of three components that matches the data within the statistical uncertainty of the measurement. As far as we can tell within the accuracy of the data, this response function is constant throughout the spectrum. This is an important advantage of MMCs over other cryogenic detector technologies, and allows us to use the response function measured accurately at 60 keV for spectral analysis at other energies.

To illustrate the MMC performance with a measurement of interest to the nuclear safeguards community, we have prepared a 120 µCi source of fissile U-233, and added 20 µCi of Pu-239 as an internal calibration standard. We have taken spectra with two MMC pixels over a period of 5 days, and compared them to HPGe spectra of the same source (Figure 8, top). As before, the response of the MMC is mostly linear, with a small but constant quadratic term (Figure 8, center). We have analyzed the U-233 / Pu-239 spectrum with the goal to accurately measure centroid energies and intensity ratios of lines with similar energies. These measurements will be important to increase the accuracy of literature values of nuclear decay data and improve the accuracy of non-destructive assay by gamma spectroscopy. In this context, we have improved the accuracy of the single gamma line of Ra by more than a factor of 50 (!) from currently 40.09 ± 0.05 keV to 40.0932 ± 0.0007 keV. Similar accuracies can be obtained



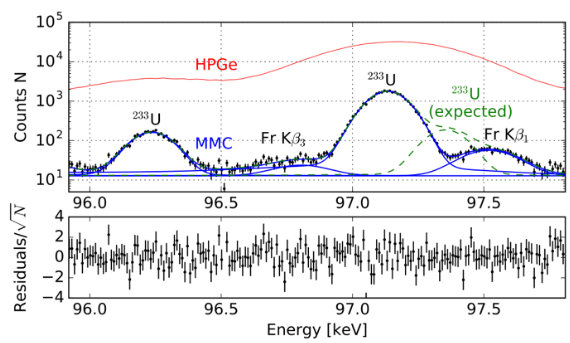


Fig. 8 (left): Gamma ray spectrum of the U-233 / Pu-239 source, taken both with a Ge detector (red) and with an MMC from Heidelberg (blue). The center panel shows the mostly linear response, with a small but constant non-linear contribution shown in the inset. The bottom panel shows the residuals from the fit, with the grey error bars indicating the magnitude of the literature errors. Note that the residuals for the two separate pixels are consistent, suggesting that the observed deviations are due to inaccuracies of the literature values. Fig. 9 (left): The 97 keV region of the spectrum can be fit (blue) to the data (black) within the statistical accuracy of the measurement (bottom panel). The fit demonstrates that the gamma emissions at 96.22 and 97.13 keV agree with the literature values, but that the postulated line at 97.37(4) keV does not exist.

for the low-energy lines of U-233. As an example, consider the 97 keV region of U-233 (Figure 9). According to the literature, it should contain three U-233 gamma-rays, although these lines cannot be resolved with a Ge detector. The high-resolution MMC spectrum of this region and the high-quality fit demonstrates that the gamma emissions at 96.22 and 97.13 keV agree with the literature values, but that the postulated line at 97.37(4) keV does not exist. We consider these values preliminary, because MMC measurements are so precise that we observe differences with some literature values, and it is currently not obvious which values are off. We plan to further assess systematic errors of the centroid energies by repeating the experiment with an MMC detector from UNM and a SQUID amplifier from STAR Cryo in FY18. Systematic differences that are observed by several pixels form different detectors in different cooldowns are likely due to errors in the literature values. Details of the measurements are described in the paper by our post-doc Geon-Bo Kim.

Outlook

We have made tremendous progress in FY17 with the completion of the first MMC gamma detector array and a measured energy resolution of 38 eV FWHM. This not only exceeds the primary performance goal for FY17, but also for FY18. We are well-positioned to scale up the photolithographic fabrication process and develop 32-pixel arrays, although UNM Ph.D. student Ruslan Hummatov, who led the fabrication effort at UNM, is ready to graduate. Our post-doc Geon-Bo Kim has been a strong addition to the team, and we will make sure that he can write several high-profile MMC publications in FY18 to strengthen the case to extend his employment and convert him to a staff scientist. The suggestion by Alain Lebrun from the IAEA to initially focus MMC applications on improving nuclear decay data is setting our direction for future work. These improved data can initially be used for higher-accuracy NDA with HPGe detectors, and subsequently for further advances of NDA with MMCs.