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Comprehensive Technology Readiness Assessment of Magnetic Microcalorimeter Gamma Detectors (January 2020)

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

Comprehensive Technology Readiness Assessment

NA-22 Project Report

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1. Project Overview

1.1 Magnetic Microcalorimeter (MMC) Project Objective

Cryogenic Gamma-ray detectors with operating temperatures below 0.1 K have been developed over the last two decades because they offer an order of magnitude higher energy resolution than conventional high-purity Ge (HPGe) detectors. This greatly reduces line-overlap and can therefore increase the accuracy of non-destructive assay (NDA) by gamma-ray spectroscopy. Among different cryogenic detector technologies, superconducting transition edge sensors (TESs) are most mature, with 256-pixel arrays with an average energy resolution of 53 eV at 97 keV having been in operation since 2012 [Bennett 2012]. While TESs provide a significant advance over HPGe detectors, they are affected by non-linearities in their response and a lack of reproducibility between cooldowns [Hoteling 2009].

Magnetic Microcalorimeter (MMC) gamma detectors are a different cryogenic detector technology that is expected to provide similarly high energy resolution without being affected by non-linearities. The goal of project LL16-MagMicro-PD2La was to develop arrays of MMC gamma-ray detectors and demonstrate an ultra-high energy resolution <50 eV FWHM. These goals were based on the predecessor proposal (LL12-MagMicro-Pd03), where we had demonstrated the feasibility of such an approach. Specifically, we had shown that:

1) MMC detector technology can be adapted for ultra-high resolution gamma-ray detection,
2) The MMC response is linear, uniform for two pixels and reproducible between cooldowns,
3) MMC gamma detectors can be used for safeguards-relevant measurements, e.g. on Pu-242.

We had summarized these results in [Bates 2016]. However, at the time we only had access to adiabatic demagnetization refrigerators, whose base temperature was limited to ~35 mK. This limited the achievable energy resolution of MMCs to ~150 eV FWHM. In addition, we had fabricated our MMC gamma-ray detectors at Heidelberg University in collaboration with the group of Prof. Christian Enss, the world's leading group for the development of MMC detectors. It was desirable to establish a source of MMCs in the US that could eventually be commercialized. Since NA-22 had also funded Prof. Stephen Boyd at the University of New Mexico for a separate effort to develop MMC gamma-ray detectors in collaboration with STAR Cryoelectronics LLC ("STAR Cryo"), one of the goals was to combine the two projects. This included the introduction of erbium-doped silver (Ag:Er) as a new paramagnetic sensor material [Boyd 2018], the development of a reliable passive heat switch for repeated thermal cycling [Humatov 2017], and geometrical modification to improve the energy resolution. Finally, the MMC detectors had to be scaled to arrays to improve the efficiency and speed of the instrument. LL16-MagMicro-PD2La therefore had three goals:

1) Purchase a dilution refrigerator with a base temperature <10 mK and adapt it for MMCs,
2) Develop 32-pixel MMC gamma-detector arrays with U of New Mexico and STAR Cryo,
3) Use MMCs from New Mexico to demonstrate an energy resolution <50 eV.

As a back-up, we have continued to collaborate with Prof. Enss' group at Heidelberg University. This ensures that we have a second source of MMCs whose response we can compare to the New Mexico detectors, and ensures regular discussions between our two efforts on the intricacies of their response.

1.2 MMC Project Description

The task to purchase a dilution refrigerator with a base temperature <10 mK and adapt it for MMC operation was relatively straightforward because it relied on proven technology. We purchased a liquid cryogen-free dilution refrigerator from BlueFors (model BF-LD400) and specified the installation of a cold finger to the side so that the MMCs can face a radioactive source next to the instrument (Figure 1). The dilution refrigerator uses a two-stage pulse-tube refrigerator for precooling to <4 K and does not require any cryogenic liquids. If no large-mass items are installed on the low-temperature stages, the instrument cools down to its base temperature <10 mK fully automated within a day (Figure 2).

For operation of MMC detector arrays, we installed 16 SQUID arrays inside superconducting Nb shielding on the 4 K stage of the dilution refrigerator, and the wiring harnesses between room temperature, the SQUID arrays and the MMCs on the 10 mK stage. We also installed 16 main amplifiers on the frame of the refrigerator for SQUID biasing, and the controllers to operate the instrument remotely. All SQUID amplifiers are commercial components from STAR Cryoelectronics.

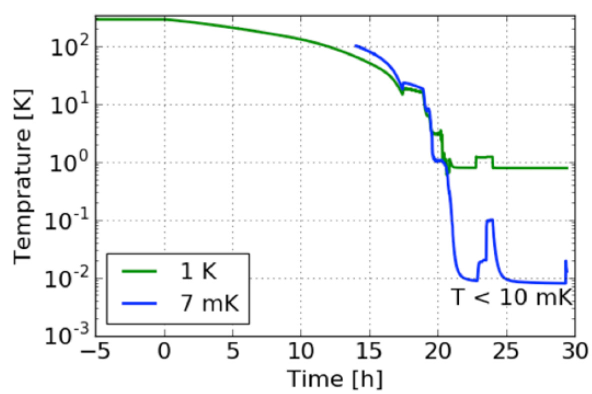
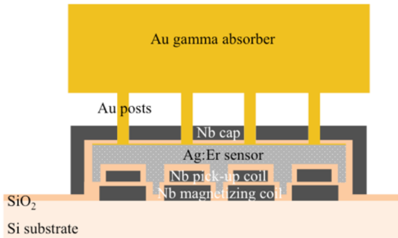


Figure 1 (left): LLNL dilution refrigerator for MMC detector operation at temperatures down to <10 mK. The MMC detector array is held behind the round cold finger at the bottom of the cryostat. The center of the flange is thinned out so that the MMC can detect gamma-rays from external sources. Figure 2 (left): The cooldown from room temperature to <10 mK is automated, does not require cryogenic liquids, and takes about ~1 day.

The development and optimization of the MMC detector arrays was the core of this project and technologically most advanced and most challenging. One difficulty is due to the fact that MMC operation requires two superconducting Nb coils in close proximity to the paramagnetic Ag:Er sensor, one high-current coil to magnetize the Er spins, and one high-sensitivity coil to pick up the magnetic gamma signals (Figure 3). This required careful optimization of the film uniformity, surface roughness and insulator thicknesses between layers. Similarly, the requirement that the Au absorber be at least ~50 µm thick and be supported on Au posts to ensure uniform energy thermalization in the absorber required advanced multi-step thick-film photolithography. Another challenge was the requirements that the Er dopants retain independent spins and thus their sensitivity at the lowest temperature, which requires that the Er not be oxidized during Ag:Er target fabrication or deposition, and that they not form Er particles whose spins start to order. Since Ag:Er sputter targets are not commercially available, this required repeated fine-tuning of our in-house Ag:Er sputter target fabrication and deposition procedures [Humatov 2018].



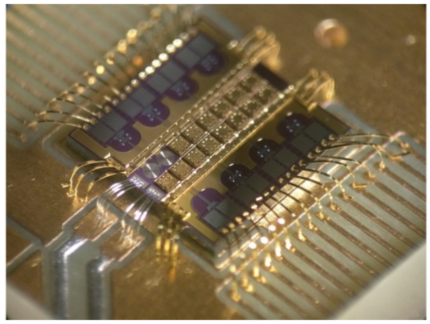
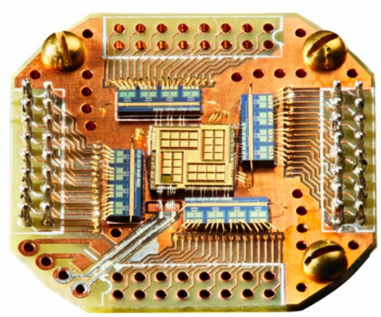
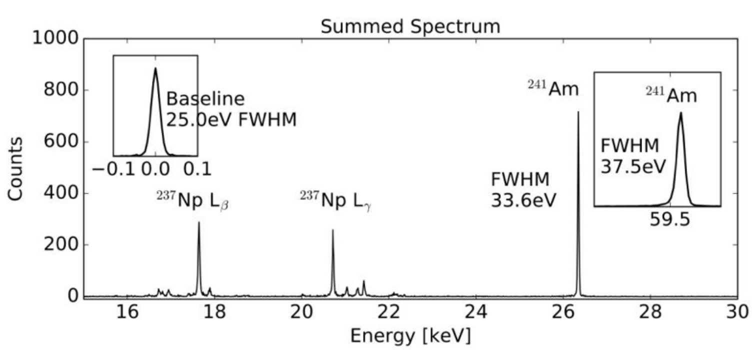



Figure 3 (right): A cross section of an MMC pixel shows the two superconducting Nb coils under the paramagnetic Ag:Er sensor, a lower one for magnetizing the Er spins and an upper one to pick up the signals. The Au gamma-ray absorber is supported on posts to ensure uniform thermalization of the energy. Figure 4 (center): 14-pixel MMC gamma detector with several different design details to compare the performance of pixels fabricated under identical conditions. Figure 5 (left): Final 32-pixel MMC gamma detector array designed at the University of New Mexico and fabricated at STAR Cryoelectronics LLC. The four central 2x4 arrays of Ag:Er MMCs with Au absorbers are read out by four SQUID preamplifiers on separate chips. The solder-covered leads on the lower left are used to apply the relatively high magnetizing current of up to 150 mA.

Our MMC detectors therefore went through several design, modeling, fabrication and test stages before settling on a few geometries that we considered most promising. In addition, we typically had implemented several detector designs in the 14-pixel arrays to test the effect of different device geometries for detectors that were fabricated under identical conditions (Figure 4). While several steps of the photolithographic detector fabrication could, other had to be newly developed and are therefore less routine. An image of a completed MMC array is shown in Figure 5. Here the central 32-pixel MMC array (one of the pixels does not have a Au absorber so that it can be used to monitor temperature drifts) is read out by 12 SQUID preamplifiers, four of which are placed on a single chip.

The MMCs have achieved an energy resolution of 38 eV FWHM at 60 keV (Figure 6) [Boyd 2018], and they have been successfully used to take high-accuracy spectra of samples relevant for nuclear safeguards (Figure 7) [Friedrich 2018]. In addition, we have measured the same U-233 / Pu-239 source with different MMCs from New Mexico and from Heidelberg and observed the *same* deviations of the



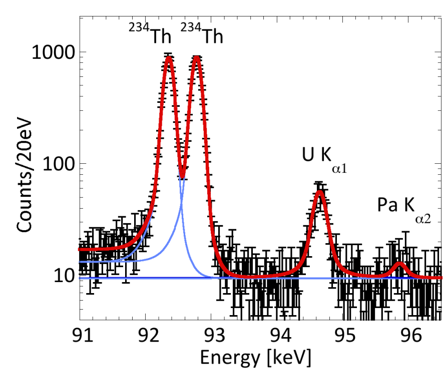


Figure 6 (left): The MMC spectrum of an Am-241 source shows a very high energy resolution of 38 eV at 60 keV, more than a factor of 10 better than HPGe detectors. This allows resolving many of the weak X-rays in the Np L manifold [Boyd 2018]. Figure 7 (right): High-resolution spectrum of a Th-234 source in the 90 keV region that is used for high-accuracy NDA of uranium enrichment [Friedrich 2018].

spectrum from literature values. This indicates that the deviations are likely due to error in the literature databases, and that MMCs have the accuracy to identify and improve these values [Kim 2018].

1.3 MMC System Description

MMC gamma spectrometers are laboratory instruments used for the non-destructive assay (NDA) of nuclear materials. The MMC spectrometer consists of the gamma detector array, the dilution refrigerator to cool it to its operating temperature of ~0.01 K, the two-stage SQUID preamplifier to read out the signals, and the controllers with the software to operate the instrument and capture and analyze the data. The refrigerator is large (Figure 1) and cannot easily be transported, so that materials typically need to be brought to the instrument for analysis. Since the MMC detectors themselves are small (Figures 4 and 5), they are mostly used for gamma-ray energies below ~130 keV, although operation up to ~250 keV possible with reduced detection efficiency.

MMC spectra (Figures 6 and 7) offer an order of magnitude higher energy resolution compared to conventional HPGe gamma detectors. In addition, their small size reduces the Compton background and increases MMCs sensitivity for low-energy signals. MMCs offer an energy resolution comparable to TES microcalorimeters, but they are much less affected by high-order non-linearities [Hoteling 2009], which greatly simplifies the calibration and addition of different spectra and the reproducibility of their response functions [Bates 2016, Kim 2018].

Applications of MMC detectors include all those where the accuracy of NDA with HPGe detectors is insufficient due to line overlap or a large Compton background, and DA by mass spectrometry is either affected by isobaric interference or too time consuming. Specific applications are:

1) Accurate non-destructive U and Pu assay in the 90 and 100 keV regions, respectively.
2) Non-destructive passive detection and quantification of isotopes that are not detectable with HPGe detectors, such as U-236 and Pu-242.
3) Isotope analysis in nuclear forensics applications where DA is too time consuming.
4) Isotope analysis of separated fuel samples where DA is too expensive, e.g. Rokkasho.
5) Independent secondary measurements of isotope ratios for important samples where DA by mass spectrometry is currently the only basis for assessment, e.g. for the IAEA SAL.
6) Increased accuracy nuclear decay data measurements for NDA and other applications.

In its current state, our MMC gamma spectrometers are at TRL5, with individual sub-systems at levels between TRL5 and 8. We have demonstrated the operation of our prototype in a typical laboratory environment, with high performance on test samples of interest in nuclear safeguards (Figures 6 and 7). Still, the detector fabrication reliability and the user-friendliness are currently not very high yet. Especially the software for data acquisition and analysis is currently custom-written and not sufficiently user-friendly to hand over to a non-expert.

2. MMC Technology Risk Summary and Readiness Assessment

2.1 Critical Technology Elements

The MMC spectrometer consists of an MMC gamma detector array, the dilution refrigerator to cool this array to its operating temperature of ~0.01 K, two-stage SQUID preamplifiers to read out the signals from the individual pixels, and the controllers with the software to operate the instrument and capture and analyze the data. These different technology elements have achieved different levels of maturity:

1) Detector: The fabrication of MMC gamma detectors involves relatively challenging photolithography, not by modern computer chip standards, but by the standards of a small high-end niche market with currently limited financial rewards. Some process steps, such as the deposition and patterning of the superconducting Nb coils and the SiO_2 insulators between them, are relatively well established, because they are used in various superconducting devices. Others, like the deposition of the paramagnetic Ag:Er sensor or the electroplating of thick Au absorbers on Au posts, are specific to MMC gamma detectors and less mature. As a consequence, the yield of our 32-pixel MMC detectors is currently only ~50%. This is acceptable for initial MMC prototypes and test results, but less so for a routine commercial instrument. (TRL-5)
2) Refrigerator: Liquid-cryogen-free dilution refrigerators with base temperatures <10 mK are by now a well-developed technology that has been commercially available from several suppliers worldwide for over a decade. They are expensive ($300k to $500k), but no longer prohibitively complicated to operate and relatively reliable, mostly due to a considerable demand for these refrigerators for quantum computing applications. (TRL-8)
3) Amplifier: Two-stage SQUID preamplifiers have also been commercially available for over a decade. But since they are used in small high-performance niche markets, mostly for scientific applications, there are only two commercial sources of these SQUID preamplifiers with sufficiently low noise to be suitable for MMC readout. One of them has temporarily stopped all sales because of a problem in their SQUID fabrication procedure, which illustrates the risk of having to rely on this technology. However, SQUID technology is relatively well understood, and the market size is sufficient so that there will likely always be at least one commercial source. (TRL-8)
4) Software: The MMC data acquisition and analysis software is currently custom-designed by each individual research group that develops MMC detectors. Most of the codes are therefore not very robust and require the presence of someone experienced with the intricacies of the code. This is partially due to the fact that typically two MMCs in a gradiometric configurations are read out with a single SQUID preamplifier, so that the raw detector output contains both positive and negative pulses that have to be separated. In addition, small temperature fluctuations cause the MMC baseline to drift over time, which needs to be corrected. These are not fundamental problems but require some effort to address. It would be desirable to develop a commercial code for MMC readout. (TRL-5)

2.2 Technology Risk Assessments

MMC gamma-ray detectors have achieved very high performance, with an energy resolution <50 eV FWHM for energies below 100 keV (Figure 6). The primary risk is whether this performance is worth the cost and the effort currently required to acquire and operate an MMC spectrometer. This risk can be addressed by either reducing cost and effort, or by increasing the value of MMCs.

The cost is dominated by the $300 to $500k dilution refrigerator, and it is not likely to decrease in the future because these refrigerators are complicated machines that required highly skilled engineers to assemble and fix. The additional cost of the MMC detectors and the SQUID readout will ensure that any MMC instrument will cost at least $500k, and likely more. This is not insignificant, but it is in line with other high-end analytical instruments. The effort to operate MMCs is currently due to a lack of maturity of the sensor and the pulse processing software. Subtle differences in MMC and SQUID characteristics so far require specific experience to optimize their operating conditions, and no MMC pulse processing software is commercially available. These risk factors can be addressed with a future focus on reliability and uniformity in MMC fabrication and on the development of a commercial well-designed software.

The value of MMCs depends on the number and importance of questions that MMCs can address that can currently not be answered at all or only at great cost. One example is the extensive role that destructive assay (DA) by mass spectrometry plays at the Rokkasho reprocessing plant in Japan. Demonstrating that NDA with MMCs can replace some of the time-consuming and costly DA at Rokkasho would increase the value of MMCs significantly. Similarly, the publication of high-profile results from MMC gamma detectors at relevant journals and conferences will raise awareness about their capabilities. In the past, our experience has been that the most valuable results from our cryogenic detectors have come out of collaboration with scientists who had heard about our work and wanted to use our technology for one of their scientific questions. Such a cross-fertilization should be encouraged through increased exchange and collaborations between cryogenic detector developers and safeguards experts.

2.3 Technology Demonstration in a Relevant Environment

Since MMC gamma spectrometers require temperatures of ~10 mK that can currently only be attained with large dilution refrigerators (Figure 1), they will almost certainly remain laboratory instruments for the foreseeable future. Radioactive sample will be brought to the MMC spectrometer for NDA, and the analysis will be performed under well-controlled conditions. Our demonstration in a laboratory setting is therefore sufficient for almost all future nuclear applications of MMCs.

Our MMC gamma spectrometers are currently at TRL-5, limited by the maturity of the pulse processing software and the lack of analyses of safeguards samples. The technology could advance to TRL-6 by improving the pulse processing software and quantifying the MMC performance on a real-world safeguards sample from a relevant product stream. Specifically, the question is under which conditions MMCs can improve relevant figures of merit, like the International Target values (ITV) of the IAEA, by how much, in what time and for which cost. This has been done for custom-designed samples, e.g. in the case of direct detection and quantification of Pu-242 in a mixed-isotope Pu sample [Bates 2016], but it should be repeated with samples relevant for safeguards or forensics. Several applications exist where the need for high-resolution gamma spectrometers is well established, e.g. [Bennett 2012, Bates 2016, Friedrich

2018, Kim 2018], but a quantification of the achievable improvements on real-world samples is mostly missing. Such a quantitative test will determine whether future developments should focus on increasing MMC energy resolution (less likely), size and detection efficiency (more likely) or count rate capabilities (most likely). For example, the IAEA has recently expressed interest in cryogenic detector technology, both TES-and MMC-based, for gamma and alpha spectroscopy, and NA-241 is currently funding the associated follow-up project. Similar collaborations with other customers are desirable.

3. Summary

Magnetic Microcalorimeter (MMC) gamma ray detectors have demonstrated very high energy resolution of 38 eV FWHM for gamma ray energies below 100 keV. This is an order or magnitude better than conventional HPGe detector and can remove most line-overlap problems in NDA by gamma spectroscopy below ~130 keV. Unlike superconducting transition edge sensors (TESs), MMCs have a reproducible and almost perfectly linear response, which greatly simplifies adding spectra from different detector pixels and reduces systematic errors in quantitative assays.

The instrument is currently at TRL-5 but could advance to TRL-6 by improving the signal processing software and using the system for the quantitative analysis of a safeguards sample provided by e.g. the IAEA. Making MMC detectors commercially viable and used more widely then depends on making the technology more reliable, specifically by increasing the device uniformity and yield, and by developing commercial software. This situation is often only half-jokingly referred to as the "valley of death" and could be addressed though the SBIR program. Our collaborators at STAR Cryoelectronics LLC are well qualified to further advance MMC development, and XIA LLC is well-positioned to develop the data acquisition system and write the software for MMC readout.

In addition, the advance to TRL-6 would be desirable to increase the visibility and showcase the value of MMC spectrometers, e.g. by placing one of them into the Safeguards Analytical Laboratory (SAL) at the IAEA, whose NDA experts have recently started to show interest in cryogenic detector technology. As a result of this LCP, we also now have an instrument at LLNL that we will use for various applications of MMCs in nuclear safeguards and science. We are confident that as the number of high-profile results increases, so will the interest in and commercial viability of this technology.

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