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Design of a Dry Dilution Refrigerator for MMC Gamma Detector Arrays

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Design of a Dry Dilution Refrigerator for MMC Gamma Detector Arrays

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Project Goal

The goal of this LCP is to develop an ultra-high resolution gamma detector 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 to replace current Au:Er sensors. The detector sensitivity will be increased by developing arrays of 32 Ag:Er pixels read out by 16 SQUID preamplifiers. MMC detectors require operating temperatures of ~15 mK and thus the use of a dilution refrigerator, and the desire for user-friendly operation without cryogenic liquids requires that this refrigerator use pulse-tube pre-cooling to ~4 K. For long-term reliability, we intend to re-design the heat switch that is needed to apply the magnetizing current to the Ag:Er sensor and that used to fail in earlier designs after months of operation. A cryogenic Compton veto will be installed to reduce the spectral background of the MMC, especially at low energies where ultra-high energy resolution is most important.

The goals for FY16 were 1) to purchase a liquid-cryogen-free dilution refrigerator and adapt it for MMC operation, and 2) to fabricate Ag:Er-based MMC γ-detectors with improved performance and optimize their response. This report discusses the design of the instruments, and progress in MMC detector fabrication. Details of the MMC fabrication have been discussed in an April 2016 report to DOE.

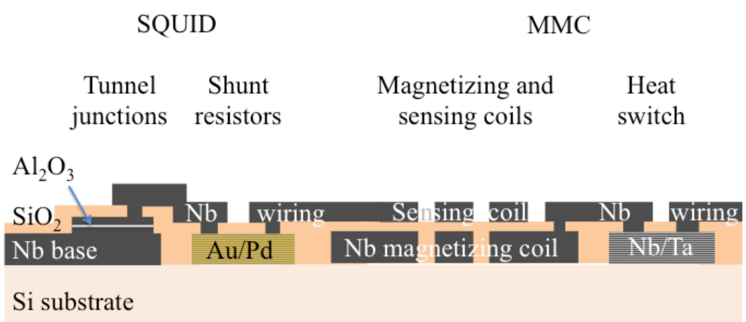
Ag:Er MMC Development (Tasks 2, 3, 5, and 6)

One goal of this project was to combine the Ag:Er sensors that UNM had developed in their predecessor proposal with the Au absorbers that LLNL had developed in collaboration with Heidelberg University. In addition, we are developing several improved device geometries that increase device sensitivity, signal-to-noise ratio and reliability. Specifically:

1) We have designed and fabricated photomasks with device layouts where the MMC detector and first-stage SQUID preamplifier are placed on the same chip (Figure 1). This allows us to eliminate the flux transformer and to use the Nb pick-up coil of the magnetic sensor to directly couple the MMC signal into the SQUID preamplifier. Since flux transformers can at best be 50% efficient, this design change can increase the signal-to-noise ratio by more than a factor of 2. We have then fabricated detector chips that contain both transformer-coupled and directly-coupled MMCs next to each other (Figure 2). Unfortunately, the SQUID fabrication process has been less consistent for our fab runs than it had been in the past. This has so far made experimental tests of the expected improvement inconclusive. We suspect that these inconsistencies are due to impurities in the oxidation process of the tunnel junction. We are currently working on identifying and fixing the underlying problem.

2) Secondly, we are fabricating MMCs based on Ag:Er sensors, in addition to the traditional Au:Er sensors (Figure 3). In the 1990s, Au:Er was chosen as a sensor material because its magnetic properties were well understood and known to be acceptable, although it was known even then that gold is not ideal because of its large nuclear quadrupole moment and thus increased heat capacity. Silver, on the other hand, consists of the two spin ½ isotopes ^{107}Ag and ^{109}Ag without a nuclear quadrupole moment, and Ag:Er MMCs will therefore have a larger signal and thus higher energy resolution because no energy is lost into the nuclear spin system. Our Ag:Er sensor is deposited from a custom-designed Ag:Er target in a dedicated sputtering chamber, and is patterned by lift-off. The fabrication works well, and will most likely form the basis for all our future MMC detectors.

3) In addition, we have integrated metallic AuPd pads into our SQUID preamplifiers to prevent the energy dissipation in the shunt resistors from heating the electrons above the cryostat temperature (Figure 1). These "cooling fins" keep the electrons in the SQUID shunts at the temperature of the cryostat and thus reduce the SQUID noise to its lowest value.



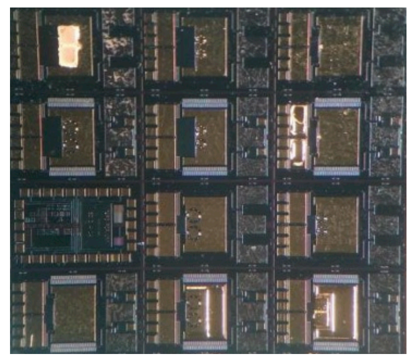


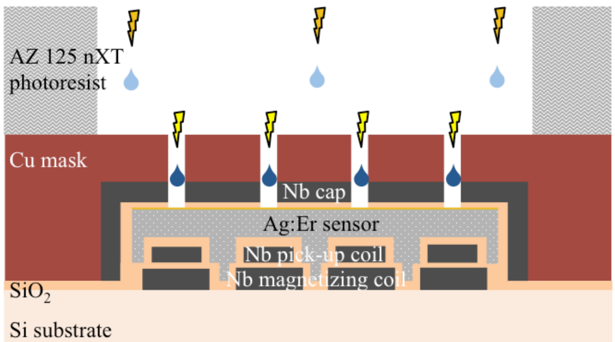
Figure 1 (right): Schematic cross section of the SQUID wafer during fabrication. The Au/Pd films serves both as a shunt resistor of the tunnel junction for SQUID operation and as a "cooling fin" for electrons. The Nb/Ta heat switch automatically loses its resistance when cooled below the critical temperature T_C = 5.5K. Figure 2 (Left): After SQUID fabrication, the 4" wafer is diced into 1 cm^2 chips for integration of the MMC detector. This chip contains different device geometries to test the expected performance improvements experimentally.

4) Also, we have added a superconducting capping layer above the Ag:Er sensor to confine the magnetic field to the sensor region and thus to increase the field at the location of the Er dopants (Figure 3). This improves the coupling between magnetic sensor and preamplifier, increases the γ-induced signal per magnetization change of the paramagnet and thus again improves the energy resolution of the detectors.

5) We have further improved the quality of the SiO_2 insulator between the Ag:Er sensor and the Nb pick-up coil (Figure 3). The thickness of this insulator determines the coupling between the MMC sensor and the SQUID preamplifier, and a higher quality of the oxide allows making this layer thinner and improve coupling. The problem in the past has been that a thin SiO_2 layer would often cause shorts, either during fabrication or during wire bonding when a small electric discharge could cause the insulator to break down.

6) We have fabricated MMC detectors with Au absorbers that are supported on the paramagnetic Ag:Er sensor by posts (Figure 4, 5 and 6). The Au posts serve as a thermal bottleneck to ensure that the gamma ray energy is fully thermalized in the absorber before heating the Ag:Er sensor underneath. The mold for the absorbers contains two different

photoresists that need to be patterned independently (Figure 3). We have encountered some unexpected difficulties with this fabrication step, as the photoresists that we have used in the past now seem to be incompatible. We are currently exploring alternative materials as photoresist, such as a Cu hard mask that can be etched away after electroplating the Au absorber (Figure 3). This process seems to work more reliably, and we have used it to successfully fabricate test chips of Au absorbers (Figure 6). We will integrate this absorber fabrication step with the rest of the process in FY17.



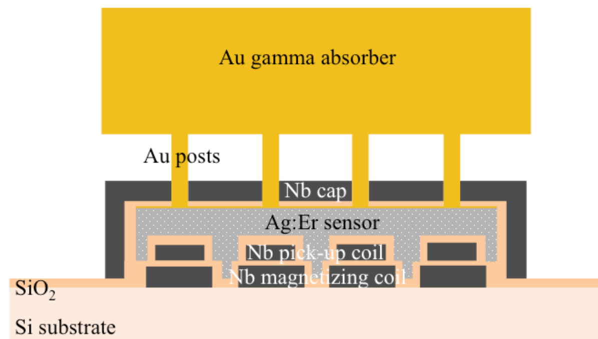
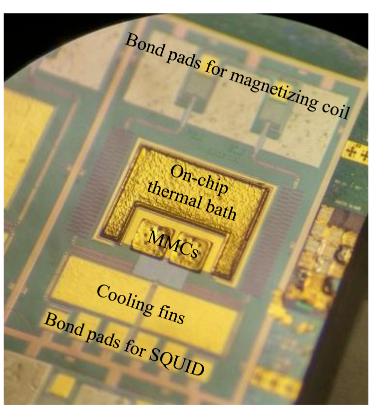
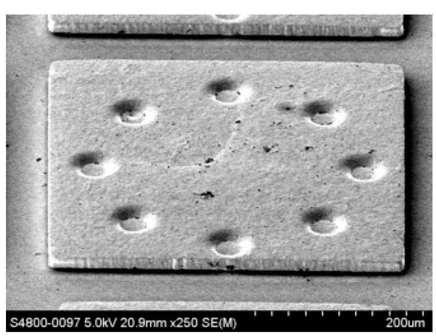


Figure 3 (left): MMC cross-section after Ag:Er sensor deposition, patterning of the Cu masks for the Au posts and the AZ-125-nXT photoresist mold for the Au absorber. The flashes and drops symbolize the acid etch to pattern the Cu and the developer to pattern the AZ-125-nXT photoresist. Figure 4 (right): Cross section of the completed MMC after electroplating the Au absorber and removal of the Cu mask and the photoresist mold.

7) For increased reliability, we have replaced the AuPd heater of our persistent current switch with a passive heat switch based on Ta/Nb with a critical temperature T_C = 5.5 K. Our field-generating loop will now automatically switch to zero resistance once the heat switch cools below T_C, rather than requiring activation with a current pulse through a resistive AuPd heater, which was prone to failure after months of operation. This new development has worked out very nicely, and we have published the results in *IEEE Transactions on Applied Superconductivity*.





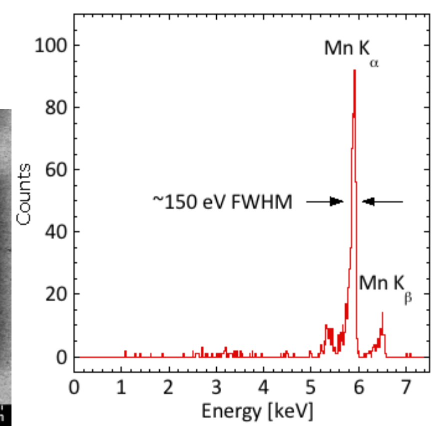


Figure 5 (left): First complete Ag:Er MMC gamma detector with electroplated Au absorber and on-chip integrated SQUID preamplifier. Figure 6 (center): Test chip of Au absorbers with posts using a Cu masks. Figure 7 (left): An Fe-55 spectrum of the first Ag:Er MMC at 50 mK shows that the basic design works. The performance of this detector will improve at T = 10 mK, and the detector design will be further optimized in FY17.

We have tested the first Ag:Er MMC with an Fe-55 source at low energies in one of our adiabatic demagnetization refrigerators at T = 50 mK (Figure 7). At these temperatures, the energy resolution is only 150 eV FWHM so far. But once the dilution refrigerator is available gives us access to temperatures down to 10 mK, the resolution is expected to improve by up to a factor of 3. In addition, electromagnetic pick-up did reduce the detector performance in these initial tests. We will also study the detector response and its noise characteristics is much more detail to determine with aspects of the detector geometry can still be improved, either for increased energy resolution or higher speed.

For an alternative source of MMC gamma detectors, we continue to collaborate with the group of Christian Enss at Heidelberg University.

Dilution Refrigerators (Tasks 1 and 7)

Dilution refrigerators exploit the fact that a mixture of liquid ^{4}He and ^{3}He phase separates at temperatures below ~1 K, with a lighter ^{3}He-rich phase on top and a heavier ^{4}He-rich phase at the bottom. Cooling is achieved by "evaporating" ^{3}He from the ^{4}He rich phase in the still, thereby forcing additional ^{3}He atoms across the ^{3}He-^{4}He interface into the ^{4}He-rich phase. This process of ^{3}He dilution is associated with a latent heat, similar to evaporative cooling of liquids, and thereby causes the temperature to decrease. Since the dilution process scales with T^3 (as opposed to evaporative cooling, whose efficiency decreases exponentially with temperature), dilution refrigerators can attain base temperatures below 10 mK. This is the temperature range needed for high-resolution MMC operation.

<u>Specifications</u>: For this project, the primary need was a base temperature below 10 mK, below 8 mK if possible. Cooling power should be as high as possible to allow a maximum number of wires to the MMC detector array, at least 10 μW at 20 mK, ideally above 15 μW. Even if a lower cooling power might have sufficed for the current project, future detector upgrades with an increased number of pixels for higher sensitivity will require additional wires and thus additional cooling power. For user-friendly operation, the dilution refrigerator should not require precooling with liquid nitrogen and liquid helium, but use a liquid-cryogen-free pulse tube refrigerator. In addition, the refrigerator should work reliably for many years without the need for a service contract. In the past, dilution refrigerators often suffered from leaks in the lines with the He^3-He^4 mixture at joints between different parts, and weld joints (rather than the more common hard solder joints) would be greatly preferred for increased reliability. Finally, the refrigerator needed to be equipped with a side port that would allow the installation of thin windows to bring radioactive sample as close to the MMC as possible.

Different companies worldwide offer dilution refrigerators with nominally similar specifications, among them Oxford Instruments, Janis Research, Leiden Cryogenics, BlueFors, and Entropy Cryogenics. We have discussed different dilution refrigerator designs with many colleagues with experience in running these instruments with cryogenic detectors or in general low-noise low temperature physics experiments. Among the suppliers, the dilution refrigerators by BlueFors are widely considered the best instruments currently on the market. They not only offer the lowest base temperature (<8 mK) and highest cooling power (>14 μW at 20 mK), they also are known for their excellent engineering and reliability, in part because they only use weld joints in the He^3-He^4 lines. We therefore purchased our dilution refrigerator from BlueFors (Figure 8).

Vibration Isolation: Good isolation of the MMC detector from vibrations of the pulse tube and external sources is essential to avoid microphonic noise and enable high energy resolution. This requires primarily that the pulse tube refrigerator is mechanically decoupled from the refrigerator, while still transferring its cooling power efficiently. For this purpose, the refrigerator will support the pulse tube on bellows below the 300 K base plate, and couple the 50 K and 4 K cold stages of the pulse tube with thick but flexible copper braids to the refrigerator to keep vibration coupling as low as possible. In addition, we have purchased an active vibration isolation system to prevent ground vibrations from coupling to the refrigerator and the signal lines. Future upgrades to improve vibration isolation are possible, but expensive and probably not required for this project. Similarly, we can install additional vibration isolation such as sound-absorbing materials around the cryostat wall and active magnetic shielding (to reduce coupling to vibrating wires) later, but decided against them at this point.

Electromagnetic shielding: Electromagnetic interference from external power sources can often dominate the electronic noise and limit the attainable energy resolution. We are therefore paying significant attention to proper grounding and shielding. The 300 K Al shell of the refrigerator serves as a Faraday cage to shield the MMC from external electromagnetic radiation, but only if it is fully electrically conducting across the interfaces between different parts of the shell. We are therefore alodining all parts to replace the natural aluminum oxide with a conducting finish ("chromate conversion coating"). In addition, all wires into the refrigerator, which break the Faraday cage, are filtered at the room temperature feedthroughs. Once the refrigerator is delivered, we will add magnetic shielding around the MMC detector array, most likely vanadium permendure or A4K at the still stage at 0.7K, plus a superconducting Nb shield at 10 mK. Should it be necessary, we can add mumetal shielding at 300K around the refrigerator later. The choice of shielding needs to be balanced with the need for low Compton scattering in the shield to maintain a low spectral background in the MMC signal. We have therefore performed Monte-Carlo simulations of the scattering contribution from the shield to the MMC spectra, and optimized the shield geometry for minimum Compton background (Figure 9). These simulations will guide us in FY17 when we fabricate and install the inner shields.

The second source of noise is from magnetic pick-up through ground loops in the circuit layout. To avoid any ground loops, the refrigerator is electronically insulated from its support frame and from the pulse tube refrigerator through plastic spacers. We have also laid a dedicated high-conductivity ground connection to the building ground, whose endpoint at the refrigerator will serve a single ground point to avoid ground loops in the readout electronics.

Future Options: We are installing several coaxial cables to the cold stage in anticipation of future multiplexing efforts to increase MMC array sizes. While not necessary for this project, the field of cryogenic detectors in moving in the direction of microwave multiplexing to read out large detector arrays, and we want to keep the option open to benefit from these developments and increase MMC sensitivity in the future.

We have also considered to purchase a load lock with a fast sample loading option for the cryostat. Although this would have been convenient for possible future experiments on alpha detection, we decided against a fast-loading option because it would have required re-design of MMC mounting and orientation, and because alpha spectrometry is not part of this project.

Similarly, we decided against installing a magnet although several future experiments may require the availability of a magnetic field. However, since the installation of feedthroughs and wiring is much easier in the initial design of the dilution refrigerator, we have added feedthroughs and high-temperature cabling for magnet wires should future projects require a magnetic field. These additions were paid from complementary LLNL LDRD funding.

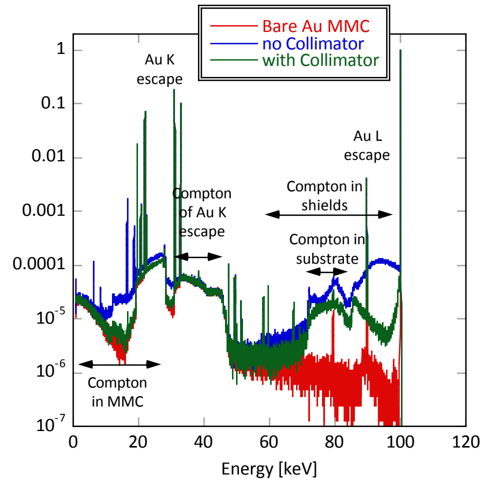


Figure 8 (left): Generic picture of a BlueFors dilution refrigerator with vibration isolation. Figure 9 (left) Monte-Carlo simulations of the Compton background from a 100 keV gamma source. We have examined different collimation and shielding geometries to optimize the design and minimize the background.

Compton Veto (Task 4)

Our initial proposal called for a scintillator operated at ~15 mK directly behind the MMC to serve as a Compton veto for forward-scattered gamma-rays. These gamma-rays set the low-energy Compton background in MMC spectra, and suppressing them by anti-coincidence vetoing increases the MMC sensitivity at low energies where many strong gamma emissions from actinides lie (Figure 10). We subsequently realized that it would be even better if the Compton veto could be made from high-purity germanium (HPGe). This would not only produce a better Compton veto, because Ge detectors have a lower detection threshold than scintillators. But a HPGe detector itself could also be used for spectroscopy at high energies where MMC lose efficiency and ultra-high resolution is less important. However, it was not obvious whether HPGe detectors could be operated with high energy resolution at millikelvin temperatures.

To test this experimentally, we have taken and existing HPGe gamma detector and modified its mounting so that it could be operated in one of our ADR refrigerators at temperatures down to 0.5 K. While the detector resolution initially remains high after cooling to 4 K and 0.5 K, it degrades over time scales of days (Figure 11). This is due to residual impurities in Ge that slowly form traps at ultra-low temperatures that can capture gamma-induced charges. The loss in resolution would be acceptable if the Ge detector only were to serve as a Compton veto. But we intend to also use it to extend the energy range of the composite detector to the MeV range, and therefore need to compensate the traps by periodically flashing the Ge crystal with light from an infrared-LED. We will work with our colleagues from the cryogenic dark matter search (CDMS), who are operating Ge detectors at 10 mK successfully, on the details of the trap neutralization in FY17.

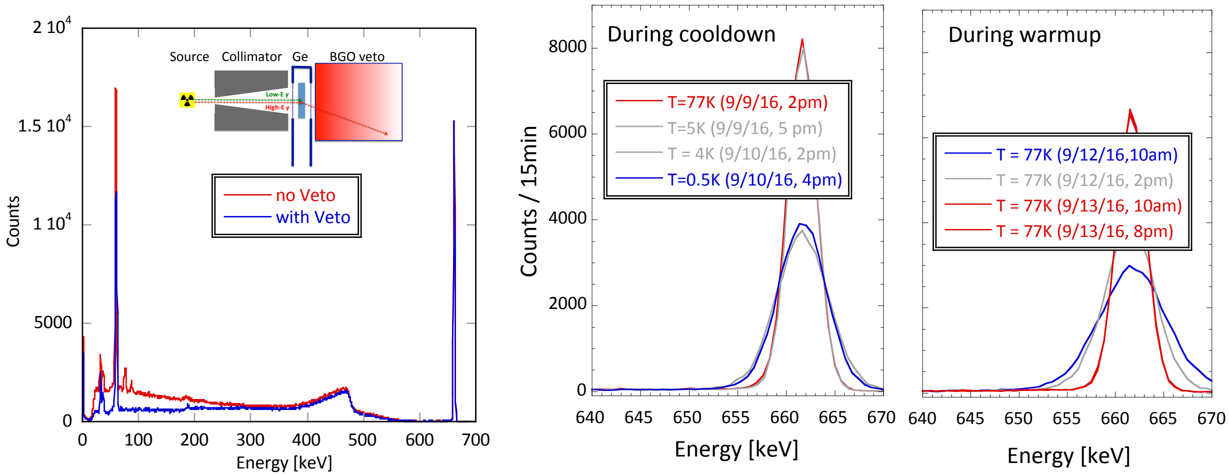


Figure 10 (left): A scintillator veto behind a HPGe detector reduces the Compton background at low energies. This demonstration experiment was performed by a CVT summer student. Figure 11 (center): The Ge detector response to Cs-137 during cooldown to 0.5K shows that HPGe detectors initially have high energy resolution even at 4K, and that the degradation does not start immediately. Figure 12 (right): During warm-up, the resolution of the Ge detector recovers only gradually, even with the Ge crystal held at 77K.

Outlook

At the beginning of FY17, all individual step of the detector fabrication process work, but need to be combined reliably to be able to fabricate MMC gamma detector arrays with high yield. The dilution refrigerator will be delivered and set up in Q1 of FY17 and allow us to test our MMCs at the temperatures down to 10 mK that they were designed for. We will then be able to study the detectors in much more detail and fine tune design and operating conditions. This will set the stage for fabricating and operating the final 32-pixel MMC array in FY18.