Device for Measuring Heat Capacities of Microcalorimeter Absorber Materials

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Abstract. We are developing a device for measuring the heat capacity of candidate absorber materials for gamma-ray microcalorimeters with the goal of finding materials with low heat capacity and high stopping power to improve detector efficiency. To date, only Sn has been effective as an absorber, and speculation is that other materials suffer from anomalously high heat capacities at low temperatures. The key component of the measurement device is a 17 mm x 17 mm low heat capacity silicon platform suspended by Kevlar fibers designed for accepting 1 g to 2 g samples, and whose heat capacity can be characterized prior to attaching a sample. The platform has a thin film Pd/Au heater deposited directly on the silicon, and a semiconducting thermometer bonded to the surface. The heat capacity is determined from $C=G\tau$, where G is the in-situ measured conductance and τ is the measured temperature decay time from a step change in applied heat. For a platform without samples, decay periods on the order of 0.3 to 0.05 seconds were measured. With samples, decay periods of several seconds are projected, allowing good resolution of the heat capacities. Several thermometers were tested in an effort to find one with the optimum characteristics for measuring platform temperatures. These included a commercial thick-film Ruthenium-oxide surface-mount resistor, a germanium NTD, and a zirconium oxy-nitride thin-film thermometer.

Keywords: Microcalorimeter; absorber; gamma-ray spectroscopy; heat capacity; thermometry.
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INTRODUCTION

For decades, High-Purity Germanium detectors (HpGe) have been the state of the art in gamma-ray energy resolution, and have therefore been the choice of spectroscopists for assaying complex plutonium-bearing materials. Recently, microcalorimeters based on superconducting transition-edge sensors (TES) have demonstrated an order of magnitude resolutions improvement over HPGe[1,2], and efforts are now underway to mature this technology into a practical laboratory instrument for nuclear safeguards and nuclear materials analysis[3].

In comparison to HPGe, microcalorimeters have substantially lower count rates, partly due to the small size of an individual microcalorimeter (~1 mm x 1 mm), and to low absorption efficiencies, roughly 20 %. The former is being addressed by developing large, multiplexed arrays[4], while the latter requires improvement in absorbers.

A good microcalorimetric absorber requires a low heat capacity, since the fundamental thermal fluctuation noise scales as $\sqrt{C}$. High atomic number Z is required for a high absorption coefficient. And since the absorption mechanism involves the ejection of an electron from an atom, the material must be able to efficiently and reproducibly convert this energy into phonons. This rules out insulators, which trap free charges whose energy goes undetected by the TES.

These properties steer absorber candidates toward high-Z superconductors, since they have low heat capacities well below the transition temperature. To date, only Sn has demonstrated high energy resolution in a microcalorimeter, despite the fact that other materials such as Ta, Mo, Re, and Pb theoretically offer comparable or better combinations of low heat capacity and high Z[5]. The reasons for the underperformance of these other absorbers are not fully understood. Typically, these absorbers display longer decay periods than expected, which can be due to extended quasiparticle relaxation times or to higher

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than anticipated heat capacities. The latter is certainly plausible since it is not unusual for materials at typical microcalorimetric operating temperatures of 100 mK to exhibit anomalous heat capacities due to, for example, small amounts of impurities.

Because of the importance of the absorber's heat capacity, we are developing a method for measuring low heat capacity samples. Results of measurements of candidate absorber materials will hopefully provide insight into the resolution degradation mechanism. In addition this device can be used for pre-screening materials or for developing processing methods such as vacuum annealing. Heat capacities of other materials used in the construction of microcalorimters, such as the patternable epoxy SU8 or Stycast 1266, can also be measured.

HEAT CAPACITY PLATFORM

The general concept for the device is to have a thermally isolated sample platform with a pre-mounted heater and thermometer. This allows measurement of the addendum heat capacity prior to the attachment of the sample. The platform is designed to measure heat capacities of a few nJ/K, which is equivalent to a few grams of superconducting Sn at 100 mK. Thermal masses on this order will have an easily measurable thermal time constant on the order of seconds. The platform is designed for measurements over the temperature range 100 mK to 500 mK.

The heat capacity platform consists of a heater and thermometer on one face of a 17 mm x 17 mm silicon chip. On the opposing face, an array of 1mm square gold pads provide a method for attaching samples such as tin with a low-melting-point solder. Low-melting-point solders generally superconduct and therefore contribute negligibly to the addendum heat capacity. Mechanical suspension is accomplished by means of four sets of Kevlar fiber strands, each threaded through DRIE-etched holes in the corners of the platform with the other ends tied to a copper frame. This frame is in turn attached to a copper base. This base allows side by side mounting of two such platforms. The platforms are covered with a copper shield to reject background parasitic radiation. The entire assembly clamps to the 50 mK stage of a standard adiabatic demagnetization refrigerator. The experimental package is shown in Figure 1.

The heater is a thin-film Pd-Au alloy strip deposited directly onto the silicon with a nominal resistance of 300 Ω. Electrical contact is made by spot welding pure niobium wire to contact pads at the ends of the heater strip. Thermometers, described below, are typically small chips with leads already attached by the vendor. The chips are bonded to the platform, and the leads are soldered to contact pads on the platform. Nb spot-welded leads are also used for final electrical contact to these pads.

The thermal conductance from the platform to the bath is primarily through the niobium leads. The Kevlar contribution was shown to be negligible by calculation and validated by varying the amount of Kevlar fiber and measuring no change in the conductance. As such, the niobium leads are thermally terminated on large copper blocks located off the platform to provide a reservoir of large heat capacity at the bath temperature. These blocks also serve as electrical terminals to connect to cryostat leads from room temperature. The final thermal conductance is of the right magnitude such that the thermal time constant with a 1g -2 g tin sample is on the order of seconds.

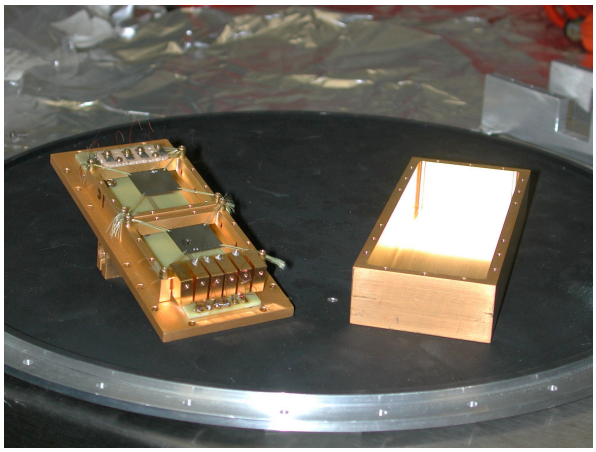

FIGURE 1. Photo of two platforms mounted on the copper base (left) along with the copper shield (right).

THERMOMETRY

We decided to use resistance thermometry for simplicity of implementation. In an effort to find the optimum combination of high-temperature sensitivity, low heat capacity, good thermal contact to the substrate, and a manageable resistance, we tested three thermometers- a commercial ruthenium-oxide (RuO) thick film surface mount resistor[6], a germanium NTD[7], and a commercial zirconium oxy-nitride thermometer[8]. Unfortunately, each thermometer lacked at least one of the desired attributes.

The particular RuO thermometer that we tested, despite the small size of a 402 package (1 mm x 0.5 mm x 0.4 mm) had an anomalously high heat capacity at low temperatures, equivalent to roughly a 2 g sample of superconducting tin at 100 mK. This heat capacity is attributed to the binder used in the RuO film itself[9]. At 100 mK, the temperature sensitivity

dR/dT of 12.7 Ω/mK was significantly lower than the 266 Ω/mK value of commercial RuO thermometers, while the resistance of 4 kΩ was roughly 20% of the commercial sensor.

The germanium NTD had a high dR/dT at 100 mK, as expected. However, the large resistance, greater than 600 kΩ at 100 mK was problematic for bridge measurements, because parasitic cable capacitance and capacitance in EMI filters become significant circuit contributions, even at the low frequencies (10 Hz - 20 Hz) of typical thermometry bridges. As a result, our experimental setup could not accurately measure such large resistances. At the kilohertz frequencies required for bare platform heat capacity measurements (see below), the parasitic capacitances are significant enough that the measurement produces an apparent positive temperature coefficient instead of the actual negative temperature coefficient. In principle, there is a temperature response, so the NTD could still be used for thermometry, but in practice this scheme is very sensitive to microphonics.

The zirconium oxy-nitride thermometer had a manageable resistance at 100 mK of 10 kΩ, and a good temperature sensitivity of 200 Ω/mK. However, this thermometer had very poor thermal contact to its own substrate. The thermal conductance, determined from the electrical resistance as a function of bridge excitation power compared to resistance as a function of platform heater drive, had a power-law temperature dependence with an exponent of roughly 4.5, suggesting electron-phonon coupling as the thermal impedance to the substrate. In contrast, for both the NTD and RuO thermometers, the measured change in resistance due to bridge excitation power was identical to that from heater power, indicating that the thermometer to silicon platform thermal conductance was significantly higher than that for platform to the bath. Despite this, a sensitivity comparison to the RuO sensor for identical self-heating temperature rises showed that the zirconium oxy-nitride sensor was superior because of its higher dR/dT.

PLATFORM CHARACTERIZATION

The heat capacity of the platform was characterized by use of the same methods that will be used to measure the heat capacity of absorber samples. The time constant τ of the exponential decay of temperature after the application of a heat pulse was measured, and in combination with a measurement of the conductance G gives the heat capacity through $C = G\tau$. The conductance G was determined from the temperature rise due to a small measured amount of power. Typical results are plotted in Figure 2. The temperature dependence obeys a power law, T^n, with n roughly 2.6. This is close to published values[10] for pure Nb of 2.7. The strong temperature dependence has the disadvantage that at the lowest temperatures, the thermometry excitation must be lowered to avoid platform self heating. A more insidious problem is that at the lowest temperatures the platform is highly susceptible to heating from parasitic electrical signals coupling into the system.

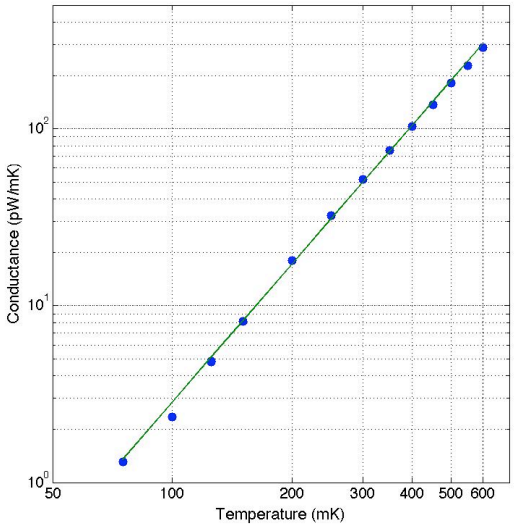

FIGURE 2. Typical conductance vs. temperature for a platform, demonstrating power-law behavior of the conductance.

A typical temperature decay in response to a heat pulse is shown in Figure 3. A lock-in amplifier was used to measure the voltage across the thermometer in response to a current excitation provided by the amplifier's internal reference source in series with a 10 MΩ resistor. Drive frequencies were necessarily high, in the kilohertz range, because decay periods for an unloaded platform were as short as 50 msec. The plot is composed of an average of several hundred decays. The linearity of the decay on a semi-log plot indicate that all components on the platform are isothermal.

Figure 4 shows the heat capacity of a typical unloaded platform with a zirconium oxy-nitride thermometer. An estimate of the heat capacity of the platform is also shown, which includes contributions from the silicon platform, bond pads, heater, thermometer with leads, and solder for thermometry leads. The heat capacity of a tiny amount of epoxy used to attach the thermometer is not included. The agreement is sufficient, considering uncertainties in the

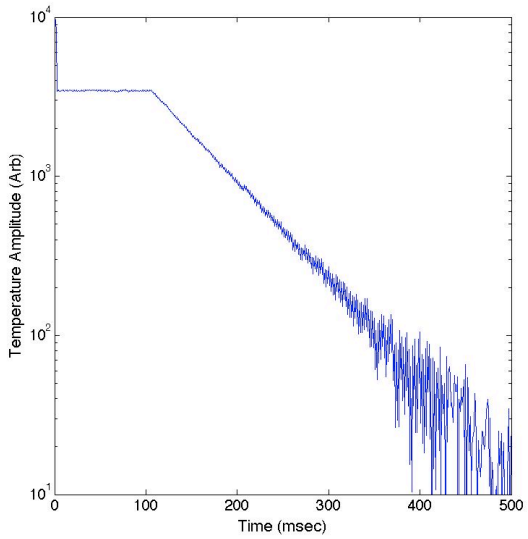

FIGURE 3. Typical decay curve in response to a step change in heat. The curve consists of an average of several hundred decays.

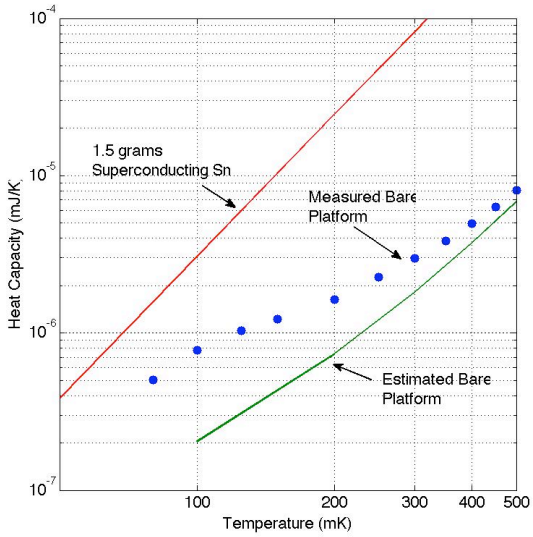

FIGURE 4. Bare platform heat capacity with a zirconium oxy-nitride thermometer.

estimated quantities, and in particular, the neglect of the epoxy, which typically has a large heat capacity near 100 mK. The heat capacity discrepancy is equivalent to a reasonable quantity of about 1 mg of a typical epoxy.

Also included in the figure is a Debye model estimation for the heat capacity of 1.5 g of pure tin. At 100 mK, the addendum heat capacity is only 25 % of the expected tin heat capacity, so any uncertainties in the addendum correction will produce no significant error in the measurement.

SUMMARY AND FUTURE WORK

The work to date suggests we have an acceptable platform for measuring the heat capacity of absorber samples, having demonstrated very good isothermalization, low addendum heat capacity, and projected decay times that are easily measured. Although the thermometers tested to date are less than ideal, they are sufficient to obtain heat capacity data. In particular, the zirconium oxy-nitride thermometer has a sufficiently low heat capacity, high dR/dT, and an easy-to-measure resistance at low temperatures to be able to perform these measurements despite the poor internal thermal contact. We are planning to initially measure presumably well-behaved samples such as high purity copper and tin to validate our technique. Subsequently, we plan to measure absorber materials that have shown anomalous behavior, such as tantalum, in addition to construction materials such as SU8 and Stycast 1266.

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