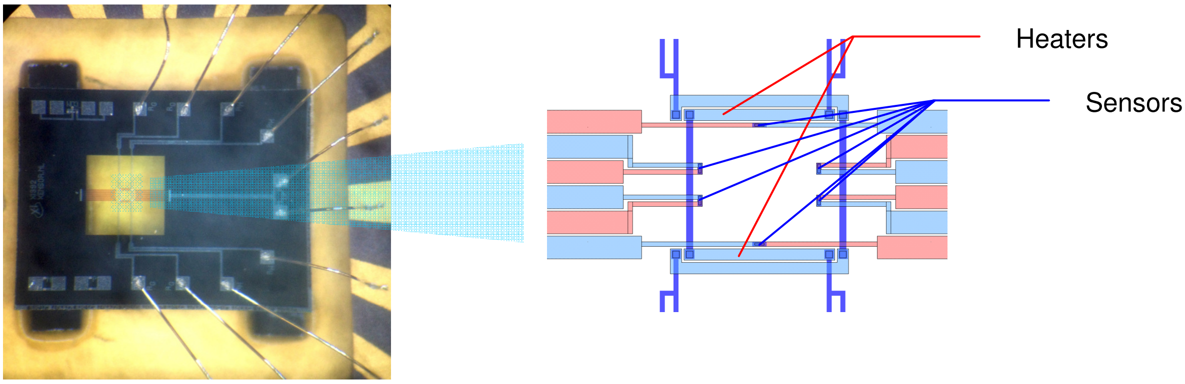
Application Note: Studying phase transitions using AC calorimetry.

The method of alternating-current (AC) calorimetry provides an efficient means to study phase transitions in small samples. It combines ultimate sensitivity, high speed of measurement and excellent thermal contact to the sample. This note presents the results of heat capacity measurements in a single crystal of a FeII spin-transition system. This is a complex magnetic material which undergoes two phase transitions at 115 and 176 K. The method is especially suited to small samples and allows the finer details of changes in thermal capacity to be studied. It is particularly useful for hysteretic phase transitions.

This method is both fast and capable of resolving details that would be very difficult if not impossible to see using other methods such as relaxation calorimetry.

The sample platform for AC calorimetry is a thin free-standing Silicon-Nitride membrane with a patterned structure that contains a heater and a temperature sensor. The method works by applying an AC current to the heater and measuring the resulting temperature oscillations (both amplitude and phase). The signal is measured by a lock-in amplifier, which makes this method very fast in comparison with adiabatic or relaxation calorimetry. Recording the heat capacity as a function of temperature across the phase transition, allows the fine structure of the transition to be resolved.

Fig.1 – Heat capacity gauge on Silicon-Nitride membrane



For this study, a single crystal with approximate dimensions 150 x 150 x 30 μm was used. The measurements were performed in a Cryogenic 5T mini Cryogen-Free Measurement System (mCFMS-5).

The temperature of the sample platform was ramped in a linear fashion between 70 and 300 K at a rate of 1 K/min and the data recorded continuously during warming and cooling.

The locally measured sample temperature slows down as the sample is warmed through the transitions. This is an indication of the latent heat being absorbed. Therefore at both transitions a first-order component is present. The magnetic transitions are coupled to structural transformations.

As the sample goes through a transition while warming, the temperature oscillations reduce, as the temperature cannot rise above T_c until the entire sample changes to the high-temperature phase. The reduction of the amplitude is caused by the peak in the observed heat capacity.

The first-order transitions are not reversible and the high-temperature phase supercools in a metastable phase below T_c. At a lower temperature there can be one or several abrupt transitions into the low-temperature phase. This happens without the latent heat affecting the amplitude of the temperature oscillations, hence the peak at the transition is suppressed or absent.

Two studies are presented below, the first on a FeII spin-transition system, the second on niobium.

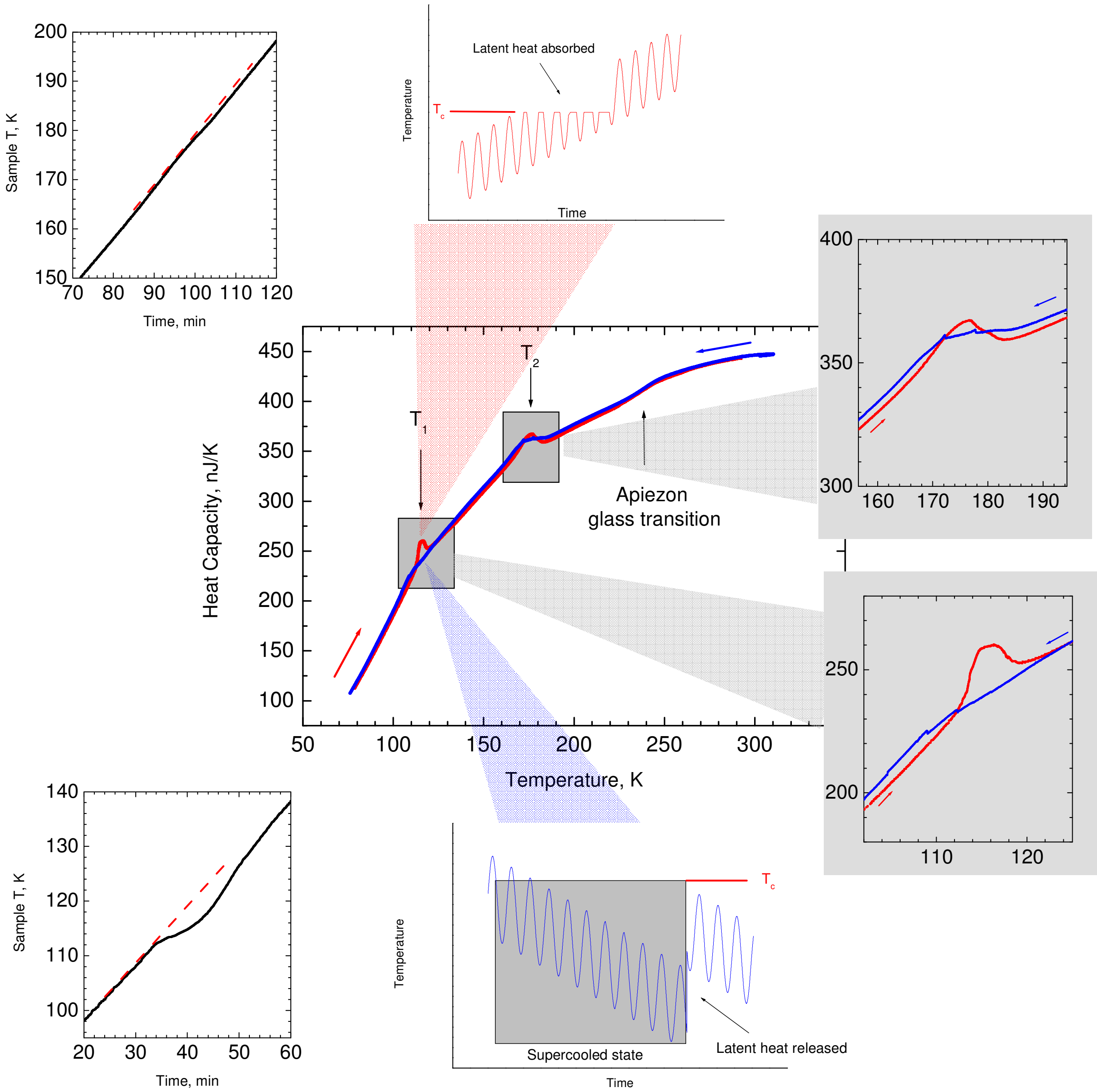


Fig.2 Central panel: temperature dependence of the heat capacity during warming and cooling cycle. Right: details of the phase transitions. Left: Sample temperature vs time, indicating that temperature ramp slows down at the transitions, due to absorption of the latent heat. Top and bottom: schematic representation of sample temperature oscillations while warming and cooling across the first-order phase transition

Heat capacity of Niobium sample.

System	Cryogen-Free measurement system, 9 Tesla
Description of measurement	AC heat capacity using silicon-nitride membrane. Frequency of temperature oscillation 256 Hz Amplitude 0.1 K approx
Measurement protocol	Continuous temperature ramp from 11 K to 6 K, than back to 11 K, repeated at different values of applied magnetic field. Temperature ramp rate was set to 0.3 K/min and cooling/warming curves were taken to verify the absence of hysteresis.
Sample	Thin plate of Niobium, 100 µm x 100 µm x 15 µm approx. Sample mass 1.3 µg. Sample mounted onto the gauge using small amount of Apiezon N; the heat capacity of Apiezon measured separately and subtracted from data. Superconducting transition point shifts to low temperature as magnetic field is applied