HEAT CAPACITY MEASUREMENTS IN PULSED MAGNETIC FIELDS

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(World Scientific, to be published)

Abstract

The new NHMFL 60T quasi-continuous magnet produces a flat-top field for a period of 100 ms at 60 Tesla, and for longer time at lower fields, e.g. 0.5 s at 35 Tesla. We have developed for the first time the capability to measure heat capacity at very high magnetic fields in the NHMFL 60T quasi-continuous magnet at LANL, using a probe built out of various plastic materials. The field plateau allows us to utilize a heat-pulse method to obtain heat capacity data. Proof-of-principle heat capacity experiments were performed on a variety of correlated electron systems. Both magnet performance characteristics and physical properties of various materials studied hold out a promise of wide application of this new tool.
I. TECHNIQUE

The 60 Tesla Long-Pulse (60TLP) magnet was recently commissioned at the Los Alamos National Laboratory. This magnet produces a flat-top field for a period of 100 ms at 60 Tesla, and for longer time at lower fields, e.g. 0.5 s at 35 Tesla. During the entire pulse, the magnetic field varies at a maximum ramp rate of \( dB/dt \approx 400 \text{ Tesla/sec} \). Together, these properties allow for the development of brand new tools to study materials in pulsed magnetic fields. Heat capacity measurement is one of these tools. We have built a probe made of plastic materials that allows us to perform heat capacity measurements at temperatures between 1.6 K and 20 K in fields up to 60 Tesla. To maximize the available experimental space a novel vacuum tapered seal was developed. The conical plug part of the joint is made out of G-10, and the matching vacuum can is made out of 1266 Stycast epoxy. The differential thermal contraction between the parts aids in producing a superfluid-tight joint. The simple construction of the joint resulted in a 16 mm diameter experimental region.

The main parts of the probe inside the vacuum can are the temperature regulated block (TRB) and the silicon heat capacity platform with sample, thin film resistive heater, and bare chip Cernox thermometer. The TRB is made of 2850 Stycast epoxy chosen for its fair thermal conductivity, and is thermally connected to the bath via two dozen thin (gauge 44 and 38) 4-inch long copper wires. The heat capacity platform is suspended with nylon strings, and electrical connections to thermometer and heater provide a thermal link as well. The resulting thermal equilibrium time constants for the TRB and the platform were measured to be on the order of few minutes. Therefore, during the magnetic field pulse, which lasts for about 2 seconds, both the TRB and the heat capacity platform can be regarded as thermally isolated from the bath and each other, and under adiabatic condition. The third time constant \( \tau_{st} \) is that of the heat capacity stage including sample, platform, thermometer, and heater. The sample’s internal thermal relaxation time constant \( \tau_{int} \) can be less than millisecond at a temperature of a few Kelvin. However it grows rapidly as the temperature is increased. At the low temperature end \( \tau_{st} \) can increase substantially due to either increase in \( \tau_{int} \) (electronic or nuclear magnetic entropy) or boundary thermal resistance between different constituents of the stage. The temperature interval between 1 K and 20 K is therefore a convenient starting point for developing heat capacity measurements in pulsed magnetic fields.

We use a heat pulse method to measure heat capacity, where a known amount of heat is delivered to the sample using a chip resistor as a heater element. The heat capacity stage must come to equilibrium both before and after the heat pulse is delivered while the magnetic field remains constant. The flat field plateau of the 60TLP magnet allows this to occur. The temperature of the stage is measured with a Cernox chip resistance thermometer, which was calibrated in both DC field up to 30 Tesla and pulsed fields up to 60 Tesla. The heat capacity of the sample is then determined as the ratio of the heat delivered to the sample to the change in its temperature. The low ramp rate of the long-pulse magnet (in comparison with short pulse, capacitively driven magnet with the total magnet pulse time of about 10 ms) reduces the unavoidable eddy current heating in metallic samples. However, during magnetic field sweep the temperature does not stay constant even in the total absence of eddy current heating, due to the magnetocaloric effect. The heat capacity stage is thermally isolated from the bath, and remains in adiabatic condition during the magnetic field pulse.
The dependence of the temperature of the stage on the magnetic field during the pulse is then given by the expression

$$\left( \frac{\delta T}{\delta H} \right)_S = -\frac{T}{C_H} \left( \frac{\delta M}{\delta T} \right)_H,$$

(1)

where \(T\) is temperature, \(H\) is magnetic field, \(M\) is magnetization, and \(C\) is the specific heat of the sample, and subscripts \(S\) and \(H\) indicate constant entropy and magnetic field, respectively. Eq. (1) is used to describe the process of adiabatic demagnetization cooling, where \((\delta M/\delta T)_H\) is negative and therefore \((\delta T/\delta H)_S\) is positive. Such a system warms as the field is ramped up, and cools during the ramp down portion of the magnetic field pulse reversibly. However, magnetization in general can also increase with temperature. The sample then would cool during the ramp up, and warm reversibly during the ramp down of the magnetic field. Below we show examples of both types of behavior.

II. RESULTS

The first heat capacity experiments in the 60TLP magnet were performed on a single crystal of metallic YbInCu\(_4\), grown from an In-Cu flux as described previously. This system undergoes a first order valence transition at 42 K in zero field. The specific volume is increased by 0.5% upon cooling through the phase transition, with accompanying rise in the Kondo temperature \(T_K\) from 25 K to 500 K. It is believed that unlike in the case of Ce, where the phase transition is described within a Kondo-collapse scenario, the valence transition YbInCu\(_4\) is driven by the band structure effects. The complete magnetic field - temperature phase diagram was obtained in DC Bitter magnets at NHMFL/Tallahassee and in capacitor-driven pulsed magnets at NHMFL/Los Alamos. This work showed that the transition can be suppressed down to \(T = 0\) K with an applied field of 34.3 Tesla.

The length of the flat top can be close to 0.5 s in the 60TLP magnet for magnetic fields less than or equal to 35 Tesla. When \(\tau_{st}\) is much smaller than the length of the plateau, a sequence of heat pulses can be delivered to it within the flat portion of the field profile, with sufficient time for the calorimeter to come to equilibrium before and after each of the heat pulses. This situation is illustrated by Fig. 1(a) for a field of 20 Tesla, where a sequence of five 10 ms long heat pulses were delivered to the heat capacity stage during the plateau of a single magnetic field pulse. The thermometer comes to equilibrium after the heat pulse is delivered well before the next heat pulse, and temperature is determined before and after each of the pulses. In this way a series of five \(C_H(T)\) data points is collected in a single ”shot” experiment, as the initial temperature for each of the heat capacity experiments is increased due to the previous heat pulse. The data from this and one zero field ”shot” is shown in Fig. 1(b). We fit the data with a sum of T-linear (electronic) and T-cubic (phononic) terms \(AT + BT^3\). For zero field we obtain \(A = 49.5 \pm 0.4\) mJ/molK\(^2\) and \(B = (0.85 \pm 0.03)\) mJ/molK\(^4\). The value of \(A\) is in excellent agreement with available data in the literature. At 20 Tesla we obtain \(A = 80 \pm 5\) mJ/molK\(^2\) and \(B = (0.81 \pm 0.07)\) mJ/molK\(^4\). The magnitude of the cubic term due to phonons is field-independent as expected. The increase of the linear term with field is likely due to scaling with magnetic field observed for various properties of YbInCu\(_4\).
Another way to increase the data acquisition rate relies on the programmable nature of the 60TLP magnet. A series of plateaus at different magnetic fields can be produced within a single experiment. Fig. 2 displays the data for one such experiment on YbInCu$_4$, with four plateaus at 25, 30, 35, and 40 Tesla, each 130 ms long. At each of the magnetic field plateaus the heat pulse is applied to the stage, and heat capacity experiment is performed. In addition, as the field was changed between 30 and 35 Tesla through the first order phase boundary, the temperature of the sample was observed to go down on the up-sweep, and up on the down-sweep, due to the magnetocaloric effect. These features are very sharp, and allow direct determination of the phase diagram of YbInCu$_4$. This is yet another complementary way to collect data using the heat capacity apparatus. It was not possible to measure heat capacity in the high field phase due to a large increase in $\tau_{st}$.

Preliminary measurements of heat capacity of UBe$_{13}$ and Ce$_3$Bi$_4$Pt$_3$ were performed up to 60 Tesla. Fig. 3 shows temperature variation of UBe$_{13}$ and Ce$_3$Bi$_4$Pt$_3$ as a function of field during magnetic field pulses to 60 T. This figure illustrates the magnetocaloric effect under adiabatic conditions of our apparatus. The temperature of the UBe$_{13}$ sample increases from 4 K up to 10 K during the ramp up, and returns to 4 K during the ramp down in a very reversible fashion. The opposite is true for Ce$_3$Bi$_4$Pt$_3$, which is colder at 60T than at 0 T. With available magnetization data it should be possible to calculate the specific heat of these compounds during the ramp portions of the field pulse, providing information in addition to the direct heat capacity measurements at field plateaus.

CONCLUSION

We have demonstrated the feasibility of heat capacity measurements in the pulsed magnetic fields provided with the 60 Tesla Long Pulse magnet at NHMFL/LANL. Direct measurement of heat capacity at field plateaus was clearly demonstrated for a variety of compounds. It appears that thermal equilibrium can be achieved in some compounds even during the field sweep, given the low ramp rates which the 60TLP magnet is capable of achieving. In this situation specific heat data can be obtained via the magnetocaloric effect from the temperature vs. field traces and magnetization data for such compounds. Other types of thermal relaxation-related experiments like thermal conductivity and Seebeck effect are also under development.

ACKNOWLEDGMENTS

This work was conducted under the auspices of the Department of Energy. It was also supported by the In-House Research Program of the NHMFL.
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FIGURES

FIG. 1. Multiple heat capacity experiments on YbInCu$_4$ during a single 20 Tesla 0.5 s long pulse. a) Dashed line - magnetic field. Solid line - voltage applied to the resistive heater. ●- Thermometer’s temperature. b) □ - specific heat at H = 0 T collected with our probe. Filled squares - specific heat at 20 T obtained from data in a).

FIG. 2. Staircase pulse shape for specific heat measurement of YbInCu$_4$. Dashed line - magnetic field. Solid line - Voltage applied to the resistive heater. ● - Thermometer’s temperature.

FIG. 3. Temperature vs. field for UBe$_{13}$ (solid lines) and Ce$_3$Bi$_4$Pt$_3$ (dashed lines) during field ramp up to 60 Tesla and back to 0. Heat pulses were applied at the 60 T plateaus for Ce$_3$Bi$_4$Pt$_3$. 
1.8 2.0 2.2 2.4 2.6 2.8

Temperature (K)

0 5 10 15

Magnetic Field

0.05 0.10 0.15 0.20

C/T (J/mol K^2)

5 6 7 8 9 10 11

H = 0

H = 20 T

fit

b)
