Thermodynamics of the magnetocaloric effect in the swept field and stepped field measurement limits

Yasu Takano[1] and Nathanael A. Fortune[2]

[1]Affiliation not available
[2]Smith College

January 29, 2021

1 Energy conservation in swept field limit

For a calorimeter sample (plus addenda) weakly thermally linked to a temperature controlled reservoir, energy conservation implies

$$-TdS = \kappa\Delta T dt + C_{\text{addenda}}\, dT \tag{1}$$

where κ is the sample to reservoir thermal conductance and addenda is the heat capacity of the actual addenda (such as the thermometer, heater, and glue or grease binding the sample to the sensors) plus the heat capacity of the sample lattice (due to phonons).

The left hand side term is the heat released by the system — which in the case of a spin system, for example, would be the heat released by the spins — when the field is changed by dH. The minus sign indicates that system entropy decreases as heat is released. Most of the released heat flows to the reservoir but some fraction heats up the addenda (to the same temperature as the system). The first term on the right hand side describes heat flow to the reservoir. The second term describes the temperature rise of the addenda. In a non-adiabatic relaxation-time or ac-calorimeter like that used in our swept-field measurements (Fortune and Hannahs, 2014), the first term dominates. In contrast, in an adiabatic measurement, the first term is negligible.

If the entropy of a system depends on temperature T and applied magnetic field H, then

$$dS = \left(\frac{\partial S}{\partial T}\right)_H dT + \left(\frac{\partial S}{\partial H}\right)_T dH \tag{2}$$

Substituting Eq. 1 into Eq. 2,

$$-T\left(\frac{\partial S}{\partial T}\right)_H dT - T\left(\frac{\partial S}{\partial H}\right)_T dH = \kappa\Delta T dt + C_{\text{addenda}}\, dT \tag{3}$$

2 Thermodynamics

We now wish to relate our expression energy conservation to the magnetocaloric effect $\Delta T(dH/dt)$, which depends on the temperature dependence of the magnetization (T), the system magnetic-field dependent heat capacity C_H, and thermal conductance κ.

From classical thermodynamics, we have the definition of heat capacity (for the spin system)

$$C_H = T\left(\frac{\partial S}{\partial T}\right)_H \tag{4}$$

and the Maxwell relation

$$\left(\frac{\partial S}{\partial H}\right)_T = \left(\frac{\partial M}{\partial T}\right)_H \tag{5}$$

For convenience, we have chosen "proper" units such that H is in tesla and M is the total magnetic dipole moment (rather than the dipole moment divided by the volume).

3 Short relaxation time limit

Substituting Eqs.4 and 5 into Eq. 3, we have

$$-C_H dT - T\left(\frac{\partial M}{\partial T}\right)_H dH = \kappa \Delta T dt + C_{\text{addenda}}\, dT \tag{6}$$

dividing through by $\kappa\, dT$, and setting $C = C_H + C_{\text{addenda}}$, we conclude

$$-\frac{T}{\kappa}\left(\frac{\partial M}{\partial T}\right)_H \frac{dH}{dt} = \Delta T + \frac{C}{\kappa}\frac{dT}{dt} \tag{7}$$

as in reference (Fortune et al., 2009). Note here that $dT/dt = \frac{d}{dt}(\Delta T)$ since the reservoir temperature is held constant.

Notice that on the right hand side of Eq. 7, $C/\kappa = \tau$, the sample to reservoir relaxation time of the calorimeter. In the short relaxation time limit, Eq. 7 reduces to

$$-\frac{T}{\kappa}\left(\frac{\partial M}{\partial T}\right)_H \frac{dH}{dt} = \Delta T \tag{8}$$

4 Stepped field limit

In stepped field (adiabatic limit) magnetocaloric measurements, the first term on the right hand side of Eq. 1 is negligible. In that limit,

$$-C_H dT - T\left(\frac{\partial M}{\partial T}\right)_H dH = C_{\text{addenda}}\, dT \tag{9}$$

Combining C_H and C_{addenda} and replacing dT, dH with ΔT, ΔH,

$$-T\left(\frac{\partial M}{\partial T}\right)_H \Delta H = C_{\text{addenda}}\, \Delta T \tag{10}$$

References

N A Fortune and S T Hannahs. Top-loading small-sample calorimeters for measurements as a function of magnetic field angle. *J. Phys.: Conf. Ser.*, 568(3):032008, dec 2014. doi: 10.1088/1742-6596/568/3/032008. URL http://dx.doi.org/10.1088/1742-6596/568/3/032008.

N. A. Fortune, S. T. Hannahs, Y. Yoshida, T. E. Sherline, T. Ono, H. Tanaka, and Y. Takano. Cascade of Magnetic-Field-Induced Quantum Phase Transitions in a Spin- 1 2 Triangular-Lattice Antiferromagnet. *Phys. Rev. Lett.*, 102(25), jun 2009. doi: 10.1103/physrevlett.102.257201. URL http://dx.doi.org/10.1103/physrevlett.102.257201.