Magnetocaloric materials

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Adiabatic demagnetisation is a magnetic cooling technology that has been used in research for many years to attain measurement temperatures below the boiling point of liquid ⁴He (4.2 K). The science of its discovery contributed to the award of a Nobel Prize in Chemistry to William Giauque in 1949.¹ In the past 15 years, research into magnetic cooling at room temperature has grown rapidly due to the promise of appliance efficiencies that could better those of conventional gas-based refrigerators and the prospect of replacing greenhouse gas refrigerants with magnetic solids. Unlike the demagnetisation of paramagnets below 4.2 K, room temperature magnetic cooling uses the entropy change at a *magnetic phase transition*.

This lecture will start by describing the classic adiabatic demagnetisation of paramagnets and will then use the material presented in the *Thermodynamics and phase transitions in magnetic systems* lecture to explore further the physics of magnetic phase transitions at room temperature. I will outline the progress towards room temperature magnetic cooling that has been made in the last 15 years and discuss some open questions that remain in the fields of fundamental physics, materials engineering and magnetic refrigerator design – questions that underline the multidisciplinary nature of magnetic cooling research.

My starting point will be a discussion of the reported values of two measures of the magnetocaloric effect. These are the adiabatic temperature change, ΔT_{ad} , and the isothermal entropy change, ΔS , caused by the application of an applied magnetic field. The vast majority of experimental research literature reports estimates of ΔS but there is sufficient ΔT_{ad} data available to plot the two quantities together on a so-called Ashby plot of material performance (Figure 1).



Figure 1: An Ashby plot of magnetocaloric material performance around room temperature in an applied field of 2 Tesla (after Sandeman²). The diamonds are experimental data. Error bars arise from the variation in magnetothermal properties across a series of compositions. The circles are theoretical maxima while the solid line is a limit on the combined values of ΔS and ΔT_{ad} for a maximum magnetic field of 2 Tesla, assuming an approximate saturation magnetisation value for Fe-Rh, (Mn,Fe)₂P-based and La-Fe-Si-based compounds.

Of the materials that possess appreciable values of both ΔS and ΔT_{ad} , there are three 3*d*-metal based compounds that are being trialled in magnetic cooling devices: (Mn,Fe)₂P-based³ and La(Fe,Si)₁₃-based⁴ compounds, and manganites.⁵ A common feature that they possess (which is not shown) is a line of first order phase transitions in (field vs. temperature) phase space that terminates in a critical point close to room temperature.

I will explore why such critical or tricritical features in phase space are of interest in magnetocaloric materials, and why the search for room temperature (tri)critical points at moderate fields (~1 Tesla) leads to the development of several areas, including

- novel methods of comparative material characterisation⁶
- finite temperature material modelling⁷
- the use of experimental and theoretical techniques to examine the role material structure at all length scales, from the atomic scale⁸ to the microstructural scale⁴
- feedback between final device design and the engineering and magnetothermal performance of magnetocaloric materials^{9,10}

Any presentation of magnetocaloric materials physics leads naturally to a discussion of other methods of solid-state cooling.¹¹ I will conclude with a brief overview of the progress and challenges in two related fields: electrocaloric¹² and elastocaloric materials.¹³

References

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