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Applications of the magnetocaloric effect in single-stage, multi-stage and continuous adiabatic demagnetization refrigerators

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ABSTRACT

Adiabatic demagnetization refrigerators (ADR), based on the magnetocaloric effect, are solid-state coolers that were the first to achieve cooling well into the sub-kelvin regime. Although supplanted by more powerful dilution refrigerators in the 1960s, ADRs have experienced a revival due to the needs of the space community for cooling astronomical instruments and detectors to temperatures below 100 mK. The earliest of these were single-stage refrigerators using superfluid helium as a heat sink. Their modest cooling power (<1 μW at 60 mK [1]) was sufficient for the small (6 × 6) detector arrays [2], but recent advances in arraying and multiplexing technologies [3] are generating a need for higher cooling power (5–10 μW), and lower temperature (<30 mK). Single-stage ADRs have both practical and fundamental limits to their operating range, as mass grows very rapidly as the operating range is expanded. This has led to the development of new architectures that introduce multi-staging as a way to improve operating range, efficiency and cooling power. Multi-staging also enables ADRs to be configured for continuous operation, which greatly improves cooling power per unit mass. This paper reviews the current field of adiabatic demagnetization refrigeration, beginning with a description of the magnetocaloric effect and its application in single-stage systems, and then describing the challenges and capabilities of multi-stage and continuous ADRs.

1. Introduction

Low temperature refrigeration is increasingly important to a growing number of scientific efforts, stemming in part from recent advances in low temperature detector technologies, but also from breakthrough developments in areas such as quantum computing. This growth is fueling the development of progressively more capable refrigerators, in the direction of lower temperature operation and higher cooling power. In addition, the rising cost of liquid helium is causing a widespread transition to the use of mechanical cryocoolers as the primary cooling system in both laboratory and space-based cryogenic systems. The challenge for low temperature refrigerators is to accommodate the higher base temperature and lower cooling power than was available from liquid helium.

Among the refrigeration technologies that can be used in the temperature ranges of interest – from several kelvin to the deep sub-kelvin regime – adiabatic demagnetization refrigerators (ADR) have many advantages that are prompting more widespread use. Based on the magnetocaloric effect, which was first observed by Langevin [4] in 1905 and proposed as the basis for refrigeration by Debye [5], ADRs are compact, robust coolers that can operate over a wide temperature range with unmatched thermodynamic efficiency. The earliest implementation, by Giauque and McDougall in the 1930s [6], was also the first to demonstrate cooling to sub-kelvin temperatures. The technique was expanded variously over the next few decades, including the use of multiple stages (Darby et al. [7]) and quasi-continuous stages (Heer et al. [8]), but these systems were hampered by the difficulty of producing the large magnetic fields required. As a result, the development of dilution refrigeration in the 1960s, with higher cooling power and continuous operation, led to a steep decline in the use of magnetic coolers.

The revival of ADRs in the 1980s traces its origin to the development of low temperature detectors for astronomy applications [9]. These were thermal detectors which benefited from the reduced heat capacity associated with cooling to (initially) ~1 K temperatures, then to temperatures in the 50–100 mK range. In the interim, magnet technology advanced markedly, both in terms of engineering current density and small wire size. The commercial availability of small diameter, multifilamentary wire makes it possible construct compact, high field (2–4 T), low current (2–4 amps) magnets that suitable for use in space missions using ADRs [10,11], as well as low cost laboratory systems.

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For space applications, ADRs have many distinct advantages, including no gravity dependence, very high thermodynamic efficiency, and an ability to control temperature without dissipating heat. The high efficiency is due to the reversible nature of the magnetocaloric effect, and is important in reducing the cooling requirements for the cryogenic system that acts as the ADR's heat sink. Ultimately this reduces the size, mass and power requirements for the instrument. Low temperature detectors tend to require very stable and precise temperature control – often at the level of 1 μK rms or better – and the unique relationship between magnetic field and temperature allows temperature to be regulated at any value of interest by feedback control of the ADR's magnet current [21,22]. Non-dissipative temperature control improves efficiency by maximizing the availability of the ADR's cooling capacity for absorbing instrument heat loads.

At the same time that ADR technology was advancing, significant progress was being made in detector technologies, fabrication techniques and arraying capabilities, resulting in vastly improved resolution and expanded array sizes, with corresponding increases in heat loads and dissipation. The consequence is a need for more capable refrigerators, in terms of both cooling power and lower operating temperature. To meet these needs, different ADR architectures are being proposed and implemented, tending now toward multi-stage configurations [12] in order to achieve wider operating range, lower heat loads on the coldest stages, lower overall mass, and even continuous operation [13], which itself significantly improves cooling power and efficiency, and reduces size and mass.

The field is rapidly expanding, under a demand for coolers that are tailored to highly specific instrument requirements, particularly space instruments, and for coolers suited to multi-purpose facilities. This paper summarizes the progress that has been made in recent years, particularly in multi-stage units, to improve the performance and utility of ADRs.

2. Magnetocaloric effect

At it most basic level, the magnetocaloric effect is a phenomenon in which certain materials warm or cool as they are exposed to increasing or decreasing magnetic fields. In paramagnetic materials, the effect originates in the interaction of an external magnetic field with the magnetic moment of unpaired outer-shell electrons, and results in the entropy having a strong dependence on magnetic field and temperature. Detailed discussions of the magnetocaloric effect may be found in the literature [14,15]. Conveniently, the entropy of a system of n (in moles) magnetic moments characterized by the quantum number J, at temperature T and in a magnetic field B_{eff}, can be expressed in analytic form:

$$S/R = n\left(x\coth(x) - (2J+1)x\coth((2J+1)x) + \ln\left(\frac{\sinh((2J+1)x)}{\sinh(x)}\right)\right)$$
$$x \equiv \mu_B g B_{eff}/2k_s T = (0.336K/T) g B_{eff}/T \quad (1)$$

where μ_B is the Bohr magneton, k_B is Boltzmann's constant, and g is the Landé g-factor for the magnetic ion. The factor $2J+1$ represents the number of possible values for the z-component of the magnetic moment. In the case of an "ideal" paramagnet consisting of non-interacting moments, these states are degenerate, and the magnetic field, B_{eff}, is simply the externally applied field. Thus we find that in zero magnetic field, the entropy per mole is $R\ln(2J+1)$.

In a real paramagnet, however, the magnetic moments interact with each other and/or the crystal field. At low enough temperature, the interaction energy causes the moments to self-align, resulting in a suppression of the entropy. In the limit of $T \to 0$ K, the entropy is suppressed to zero.

It has become common to account for these interactions by defining a background field, b, that is rms averaged with the applied field, B, such that:

$$B_{eff} = \sqrt{B^2 + b^2} \quad (2)$$

In practice, using a constant value for b will reproduce the *general* temperature dependence of entropy, but for purposes of optimizing "salt pills" (the structure that contains the magnetic refrigerant in an ADR), a more accurate representation is often needed. From measurements of entropy at zero field, for example, one can compute values of b that exactly reproduce the temperature dependence of the entropy. The functional form is difficult to justify on theoretical grounds, but we have found that an exponential of the form

$$b(T) = b_0(1 - e^{-(T/T_0)^{\alpha}}) \quad (3)$$

can reproduce the zero field entropy over a wide temperature range (0–5 K) to within a few percent.

Fig. 1 shows the zero-field entropy curves for some of the most common magnetic refrigerants use in ADRs, and Table 1 summarizes their relevant parameters, including coefficients for $b(T)$. The Neel temperature, at which the material enters an ordered magnetic state, shows a clear correlation with J and magnetic ion density. A stronger magnetic moment (higher J) and closer ion spacing (higher density) both tend to strengthen nearest neighbor interactions, and raise the temperature at which internal interactions begin to affect the entropy. The upshot is that low temperature refrigerants must be relatively dilute and have lower spin. As a consequence, they have relatively low entropy (or heat) storage capacity, and as lower temperature systems are built, increasing care must be exercised to limit the heat loads reaching the coldest stage.

2.1. ADR cycle – Single-stage system

From Eq. (2), we see one of the most important features of the magnetocaloric effect in paramagnets: the entropy depends only on the *ratio* of magnetic field and temperature, B_{eff}/T. Under adiabatic conditions, a change in magnetic field produces a proportional change in temperature. Equally important, the change in temperature is completely reversible and involves no dissipative processes. It is therefore straightforward to construct a refrigeration cycle

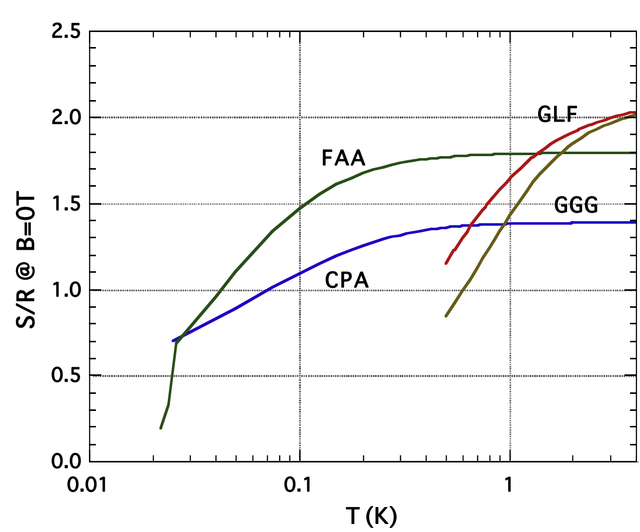

Fig. 1. Zero-field entropies for common magnetocaloric materials used in low temperature ADRs [16–18].