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WHICH OBEYS THE THIRD LAW

AUTHOR(S): J. C. Bonner, BNL
J. D. Johnson, T-4

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EXACT THEORY OF MAGNETIC COOLING FOR A SYSTEM
WHICH OBEYS THE THIRD LAW*

J. C. Bonner†
University of Rhode Island
Kingston, RI 02881 USA

and

J. D. Johnson
Los Alamos Scientific Laboratory
Los Alamos, NM 87545 USA

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Abstract

An exact solution has been developed for a reasonably realistic magnetic model known to obey the third law. Results indicate that 1-D systems are actually more efficient coolers than 3-D systems and that adiabatic demagnetization of an antiferromagnet down to the (upper) critical field is a very powerful cooling process.

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†Work performed while at Brookhaven National Laboratory, Upton, New York, 11973 USA.

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The study of magnetic cooling and the search for new magnetic thermometers and refrigerators has not seen much progress in recent years. The extant theories of magnetic cooling are either unrealistic at low temperatures because they fail to take account of spin-spin interactions (classical and quantum paramagnets) or actually disobey the third law of thermodynamics (classical paramagnet and antiferromagnetic Ising models and mean field theories).

The time seems ripe for a more sophisticated, statistical mechanical approach, and, accordingly, a solution has now been worked out for a well-known and reasonably realistic model (the spin- $\frac{1}{2}$, nearest-neighbour, one-dimensional (1-D) Heisenberg-Ising model). This solution is exact in the sense that the problem is analytically reduced to the solution of an infinite set of coupled, non-linear integral equations which are then treated numerically to an arbitrary degree of accuracy [1,2]. This numerical approach is supplemented and supported by exact analytic results in cases of special interest. The model is known to obey the third law of thermodynamics [2,3].

The results obtained so far include an extensive determination of the entropy isotherms, isentropes (cooling curves) and specific heat as a function of temperature and applied magnetic field. From these quantitative results, computations can then be made [2] of the relative cooling efficiency over different ranges of applied

magnetic field (a) for varying degrees of anisotropy and (b) for the antiferromagnet in comparison with the corresponding ferromagnet. A plot of the ^{4.4: ferromagnetic} ~~isentropes~~ ~~(cooling curves)~~ for intermediate uniaxial anisotropy is shown in Figure 1. The anisotropic system is characterized by two critical fields, H_{c1} and H_{c2} . It is amusing that four different types of cooling are possible: (1) cooling by adiabatic magnetization to the lower critical field H_{c1} , (2) cooling by adiabatic demagnetization from a field H intermediate between the two critical fields, i.e. $H_{c2} > H > H_{c1}$, down to H_{c1} , (3) cooling by adiabatic magnetization from an intermediate field to the upper critical field H_{c2} and, (4) cooling by adiabatic demagnetization down to the upper critical field H_{c2} . It might be noted that this multiple cooling phenomenon is not only a property of systems with uniaxial anisotropy: very similar cooling behaviour is seen in the case of the weakly-interacting magnetic cluster (dimer) system $\text{Cu}(\text{NO}_3)_2 \cdot 2.5\text{H}_2\text{O}$ [4], where the spin Hamiltonian is effectively isotropic (Heisenberg) in type. However, in the Heisenberg-Ising system described here, H_{c1} tends to zero as the isotropic, Heisenberg limit is approached, leaving a single critical field $H_{c2} = H_c$.

Of the four cooling processes observable in Figure 1, clearly the most efficient is cooling by adiabatic demagnetization down to the upper critical field. (This is actually the analogue of cooling

an ideal paramagnet down to its critical field $H_c = 0$). The cooling efficiency (of process (4)) for the antiferromagnet improves as the anisotropy increases. The reverse is true in the case of the ferromagnet. When a comparison is made of the antiferromagnet at its upper critical field ($H_c = H_{c2}$) with the ferromagnet of corresponding anisotropy ($H_c = 0$, always), the antiferromagnet turns out to be a strikingly better cooler. For the antiferromagnet, the entropy decreases to zero rather slowly as the temperature T decreases, as a power law ($T^{1/2}$). The entropy of the ferromagnet, by contrast, vanishes exponentially fast. The reason is, of course, that the uniaxial ferromagnet at $H = 0$ always has a gap between the ground state and the first excited states (which decreases as the anisotropy decreases, vanishing in the isotropic limit [5]) whereas the antiferromagnet at $H = H_c$ (H_{c2}) has no gap. To be useful in attaining low temperatures, a system should have as large an entropy as possible down to as low a temperature as possible. The antiferromagnet is clearly much superior in this respect. It is interesting to note that by ^{similar} ~~the same~~ argument, a 1-D antiferromagnet is also superior to a higher-D antiferromagnet, since the critical entropy is expected to behave as $T^{D/2}$, where D is the dimensionality. Of course, ultimately weak interchain interactions cause a quasi-1-D real system to cross over to 3-D behaviour at low enough temperatures, but with relatively minor effect on the argument above. The existence of a phase transition is not important in connection with magnetic cooling [6].

The practical consequences of the foregoing discussion are as follows. In a search for an efficient new magnetic cooler, it appears preferable to examine first the ranks of the 1-D rather than the 3-D magnets, and it appears also that antiferromagnets are preferable to ferro- (para-) magnets. Further, the cooling salts in common use, such as cerium magnesium nitrate, are magnetically diluted to remain paramagnetic down to very low temperatures. This reduces the magnetic specific heat available for refrigeration. A (quasi-) 1-D system may actually be less magnetically dilute than the common paramagnets. Accordingly, cooling experiments are in progress on quasi-1-D, spin- $\frac{1}{2}$ copper salts [7]. The one disadvantage of antiferromagnets is that their (upper) critical field may be rather large in comparison with the maximum field obtainable with the currently available superconducting magnets (100-120 kOe). The (upper) critical field is related to the intra-chain exchange integral J , which should be as small as possible.

Unfortunately, a 1-D nuclear spin antiferromagnet is not available, but magnetic cooling by both adiabatic magnetization and demagnetization should be possible for the BCC nuclear spin- $\frac{1}{2}$ antiferromagnet, solid ^3He .

REFERENCES

- [1] J. D. Johnson, Phys. Rev. A, 9 (1974) 1743.
- [2] J. C. Bonner and J. D. Johnson, in preparation. A much more detailed, mathematical presentation of which the qualitative highlights are reported here.
- [3] J. C. Bonner and M. E. Fisher, Proc. Phys. Soc., 80 (1962) 508.
- [4] M. W. van Tol, K. M. Diederix and N. J. Poulis, Physica 64 (1973) 363. Jill C. Bonner and Simeon A. Friedberg, in Phase Transitions 1973 (Proceedings of the Conference on Phase Transitions and their Applications in Materials Science), H. K. Henisch, R. Roy and L. E. Cross, eds. (Pergamon Press, 1973) p. 429.
- [5] J. D. Johnson and J. C. Bonner, unpublished work.
- [6] J. C. Bonner and J. F. Nagle, Phys. Rev. A (1972) 2293.
- [7] S. A. Friedberg, private communication.

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Caption to Figure 1

Cooling isenuropes for a 1-D antiferromagnet with intermediate anisotropy, plotted in terms of reduced temperature KT/J and magnetic field gSH/J . J is the 1-D nearest-neighbour exchange constant.