COOLING BY ADIABATIC MAGNETIZATION

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We report 5.5° K cooling at 20° K in the ferrimagnet ytterbium iron garnet accompanying an increase of magnetic field. The cooling abruptly ceases at the phase boundary between a collinear and canted sublattice configuration. A cooling of 10° K at 18° K and 3.7° K at 6° K was observed in dilute ytterbium iron garnet.

We have observed large "adiabatic magnetization" cooling of the ferrimagnet ytterbium iron garnet by turning on a magnetic field. The largest temperature reduction which we obtain is about 100 times larger than any previously reported adiabatic magnetization cooling.¹ The idea is simple.² In the rare-earth iron garnets the exchange interaction between the two iron sublattices is very large compared with the molecular field H_m coupling the rare earth to the iron. Suppose that the net moment of the two iron sublattices (hereafter considered a single sublattice with moment \vec{M}_{Fe}) exceeds that of the rare earth, whose moment is \vec{M}_{R} . Then in a small external field H_0 pointing up, $\widetilde{M}_{Fe}(T)$ points up, and $M_R(T,H)$ points down. The net effective field acting on the rare earth is then $-H_m + H_0$. Under appropriate conditions,² as H_0 is increased to H_m , the net field on the rare earth becomes zero, the rare earth becomes completely disor-



FIG. 1. The change in temperature of ytterbium iron garnet with field. As the field is increased the temperature falls from near 22°K to 16.5°K. A decrease in the field returns the temperature to its initial value. No temperature change is observed above $H_I = 65$ kOe. Above this field the system is in the angled phase.

ganized, and thus the rare-earth spin system absorbs heat from the lattice. A further increase in H_0 orders \vec{M}_R along \vec{M}_{Fe} , and at $H_0 = 2H_m$ the crystal temperature returns to its initial value. This is the case when both sublattices remain aligned with H_0 . Under different conditions² there is a lower critical field H_1 , below which cooling occurs as described above. Between this lower field and an upper critical field H_u the rare-earth sublattice rotates upward coherently toward H_0 (i.e., both \overline{M}_R and \overline{M}_{Fe} form angles with H_0). Above H_u the oriented rare-earth moment is increased by H_0 . In this situation the temperature of the crystal falls as H_0 rises to H_1 , is stationary for $H_1 \leq H_0 \leq H_u$, and rises for $H_0 > H_u$. Figure 1 shows the behavior of Yb₃Fe₅O₁₂ in the temperature range 15-25°K. The phase boundary between the collinear and angled spin configurations is traversed at $H_1 = 65$ kOe where an abrupt termination of cooling is observed. In a different sample, with composition Yb_{0.9}Y_{2.1}- Fe_5O_{12} , we have observed cooling from 18 to 8°K and from 6.6 to 2.9° K in a field of 110 kOe.

Molecular field theory describes quite satisfactorily the phase boundaries of the angled region and the temperature and field dependence of the magnetization over the entire phase diagram. With appropriate substitutions of nonmagnetic ions one can adjust the system to produce maximum cooling in other temperature ranges of interest, say below 4.2°K, and to conform most efficiently to the available laboratory field.

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¹J. H. Schelleng and S. A. Friedberg [J. Appl. Phys. <u>34</u>, 1089 (1963)] observed adiabatic magnetization cool-

ing of the antiferromagnet MnBr₂·4H₂O. They reported $\Delta T \cong -0.125^{\circ}$ K from $T_{i}=1.75^{\circ}$ K. K. P. Belov, E. V. Talalaeva, L. A. Chernikova, and V. I. Ivanovskii, Zh. Eksperim. i Teor. Fiz. – Pis'ma Redakt. 7, 331 (1968),

have recently observed $\Delta T \cong -0.05^{\circ}$ K from $T_{i} \cong 290^{\circ}$ K in gadolinium iron garnet.

²A. E. Clark and E. R. Callen, J. Appl. Phys. <u>39</u>, 5972 (1968).

LOW-TEMPERATURE PHASE TRANSITION IN CERIUM MAGNESIUM NITRATE

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Magnetic-susceptibility measurements on cerium magnesium nitrate covering frequencies from dc to 200 Hz indicate that it undergoes an antiferromagnetic phase transition in the millikelvin range.

We report some recent observations on the lowtemperature magnetic phase transition in cerium magnesium nitrate [Ce₂Mg₃(NO₃)₁₂·24H₂O, commonly abbreviated CMN which lead to a new interpretation of the nature of the transition. Earlier reports on this transition were made by Mess et al.^{1, 2} and by us,³ but there remained some doubt as to whether it was a ferromagnetic or an antiferromagnetic transition.⁴ Our most recent measurements strongly suggest that it is in fact antiferromagnetic rather than ferromagnetic as suggested by Mess et al. One is especially interested in this point because the CMN transition occurs at a lower temperature than any heretofore observed, with the exception of nuclear magnetic transitions. Furthermore, CMN is widely used as a thermometer at temperatures as low as 1.8 mK,^{5,6} which is within the transition region, and consequently the details of the transition bear directly on the thermometry problem.

We have investigated the transition by means of measurements of the susceptibility in the frequency range 0-200 Hz as a function of time (as the sample warmed up following demagnetization) and of the initial entropy. The measurements reported here were performed on single-crystal spheres of 20 mm diam. A most striking feature of these measurements is the observation that the susceptibility goes through a maximum as the sample warms up. This was true over the entire frequency range from dc to 200 Hz provided that the initial entropy S/R < 0.1. Similar results were obtained with powder spheres and powder cylinders (of diameter and length both equal to 24 mm).

The same effect was exhibited in another type of measurement. If the salt is adiabatically demagnetized from different initial magnetic fields, and hence at different but constant entropies, the susceptibility attained when the field reaches zero also shows a maximum when plotted against the entropy.

The powder samples were compacted by adding a little water to them at room temperature and then cryopumping off the excess. This resulted in a solid compacted cylinder in which the individual grains were cemented together. The crystal spheres were made from one or two large single crystals. The sample was cooled by single-stage adiabatic demagnetization from helium temperatures (1-1.2 K) and initial fields up to 94 kG. A guard salt of chrome potassium alum was mounted above the sample and thermally isolated from it. A "coil-foil"⁷ heat shield, thermally bonded to the guard salt, surrounded the sample but was not in thermal contact with it. A set of coils and a mutual-inductance bridge⁸ were used to measure the in-phase (χ') and out-of-phase (χ'') components of the susceptibility in the frequency range between 1.5 and 200 Hz. At lower frequencies, between 0.08 and 1.5 Hz, the amplified and partially balanced signal from the secondary coils was displayed on a two-pen recorder together with the primary signal. The dc measurements were made with an integrating amplifier magnetometer (Magnemetrics Type MF-1) and the susceptibilities obtained from the initial