

## Letters to the Editor

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### Rotation Cooling in Cerium Magnesium Nitrate\*

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**C**OOLING by rotation was suggested by Bogle, Cooke, and Whitley<sup>1</sup> and has been applied in the present experiments to cerium magnesium nitrate. In this salt, at sufficiently low temperatures, the only level occupied by the cerium ion is a Kramers doublet. There is no hfs. In a magnetic field  $H$ , the level splits by  $g\beta H$ . Cooke *et al.*<sup>2</sup> found  $g$  to be symmetric about the trigonal axis with  $g_{\parallel} = 0.25 \pm 0.05$  and  $g_{\perp} = 1.84 \pm 0.02$ . For an angle  $\theta$  with the axis  $g$  is defined by

$$g = (g_{\parallel}^2 \cos^2\theta + g_{\perp}^2 \sin^2\theta)^{1/2}. \quad (1)$$

If the sample is isothermally magnetized, the entropy removed is a function of the quantity  $x = g\beta H/2kT$ . In a subsequent isentropic process,  $x$  is constant as long as  $g\beta H$  is much greater than the dipole-dipole interactions. Thus in ordinary adiabatic demagnetization  $T$  decreases directly with  $H$ . On the other hand, cooling is produced with  $H$  constant in an isentropic rotation since in this case  $T$  is proportional to  $g$ .

The experiments were performed on a 2.8-g single crystal of  $\text{Ce}_2\text{Mg}_3(\text{NO}_3)_{12} \cdot 24\text{H}_2\text{O}$  oriented so that the trigonal axis could make an arbitrary angle with the strong field. The  $g$  value parallel to the measuring coils was  $g_{\perp}$ . Extremely rigid mounting was necessary because of the high torques at intermediate angles. After isothermal magnetization, the field was reduced to a value which permitted the guard rings to absorb the residual gas and reduce the conduction heat leak. The magnetic temperature  $T^*$  was then obtained ballistically as a function of angle. In order to obtain the Kelvin temperature  $T$ , it was necessary<sup>3</sup> to correct the magnetic temperature only for the nonspherical shape of the sample and for paramagnetic saturation. We then obtained

$$T = (T^* + \Delta)x^{-1} \tanh x, \quad (2)$$

where  $\Delta = 2.5 \times 10^{-3} \text{ K}^\circ$ .

The results of the rotation experiment for three magnetic fields are shown in Fig. 1. If interactions may be neglected,  $T$  is proportional to  $g$ , so that the ratio

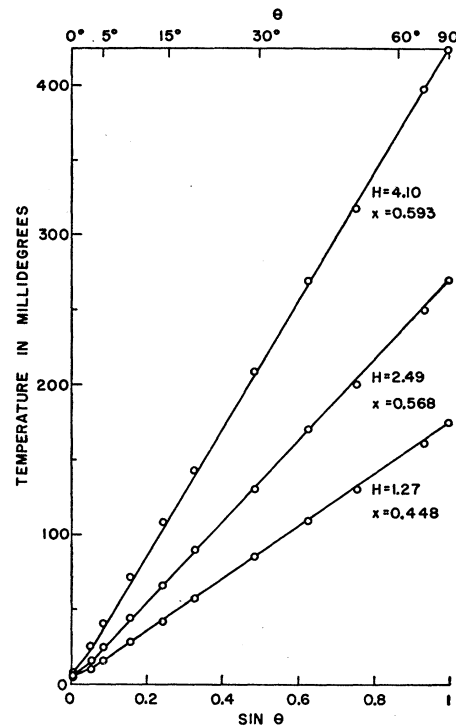


Fig. 1. Absolute temperature of the crystal as a function of the sine of the angle of the magnetic field with the trigonal axis. Each solid curve was calculated using Eq. (1) from the values of  $x$ ,  $H$  (in kilogauss), and  $T_{90}/T_0$  characteristic of the curve.

$T_{90}/T_0$  of the temperatures at  $90^\circ$  and  $0^\circ$  is  $g_{\perp}/g_{\parallel}$  if the crystal is perfectly aligned. The experimental results show that  $T_{90}/T_0$  is 57 at  $H=4.10$  kilogauss, 50 at  $H=2.49$  kilogauss, and 33 at  $H=1.27$  kilogauss. The ratio changes with  $H$  as a result of the interactions, so only a lower limit may be placed on  $g_{90}/g_0$ . Susceptibility measurements gave a Curie constant of  $(4.1 \pm 0.1) \times 10^{-4} \text{ g}^{-1}$ , in agreement with Cooke,<sup>2</sup> from which was calculated  $g_{\perp} = 1.77 \pm 0.04$ . Based on this value of  $g_{\perp}$ , the upper limit on  $g_{\parallel}$  is then 0.031. However, the data are also consistent with  $g_{\parallel} = 0$  and a misalignment of the crystal of about  $1^\circ$ . Since  $g_{\parallel}/g_{\perp} \ll 1$ , one expects  $T \propto \sin\theta$  over a wide range of angle as shown in Fig. 1. The slopes of the three straight lines are internally consistent. The foregoing value of  $g_{\parallel}$  is in disagreement with that given by Cooke.<sup>2</sup> On the basis of Judd's<sup>4</sup> theory, the ground state of the cerium is a linear superposition of a number of  $|J, J_z\rangle$  states. It is remarkable that a cancellation occurs such that  $g_{\parallel} \approx 0$ .

Rotation cooling does not itself permit lower temperatures to be reached than were heretofore attainable. However, it does have the advantages that a low temperature is produced in a strong magnetic field and that the specific heat remains constant during rotation. Moreover, the initial field may be chosen so that the specific heat has the maximum value of  $0.439 k$  per ion. Most rare earth and many iron group salts have sufficient magnetic anisotropy to make them useful for

rotation cooling. It is possible that they would be useful working substances for a magnetic refrigerator using a rotating magnet.

Work is in progress to extend the measurements to other crystals and to measure the effect of rotation on the nuclear resonance of the protons in the water molecules of the cerium magnesium nitrate.

We are indebted to Mr. H. R. Hart, Jr., for growing the crystal.

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<sup>1</sup> Bogle, Cooke, and Whitley, Proc. Phys. Soc. (London) A64, 931 (1951).

<sup>2</sup> Cooke, Duffus, and Wolf, Phil. Mag. 44, 623 (1953).

<sup>3</sup> J. M. Daniels and F. N. H. Robinson, Phil. Mag. 44, 630 (1953).

<sup>4</sup> B. R. Judd, Proc. Roy. Soc. (London) A232, 458 (1955).

## Changes in Thermoelectric Power of Copper with Cold Work at Liquid Nitrogen Temperature

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CHANGES in the thermoelectric power of copper were measured after the metal had been plastically deformed at the temperature of liquid nitrogen. The plastic deformation consisted in permanently elongating a wire specimen, having a diameter of about 0.001 inch and a length of 9 inches, which was initially annealed above the recrystallization temperature. Figure 1 shows a typical graph of the thermoelectric power at successive stages of permanent elongation. For points above the axis, the electrons flow from the reference copper wire to the cold-worked specimen at the cold junction. During each measurement the tensile stress was reduced to zero in order to eliminate the effects that elastic deformation would have upon the thermoelectric power.<sup>1</sup> This elastic deformation effect on the thermoelectric power in copper at the temperature of liquid nitrogen has a coefficient of about 12 microvolts per degree centigrade per unit strain and has a polarity which makes the electrons flow to the reference copper from the cold-worked specimen at the cold junction with the application of simple tension. The experimental uncertainty associated with each point in Fig. 1 is sufficiently small that the successive intervals of rising and precipitously falling magnitudes are entirely real. At point "A," the specimen was allowed to remain at room temperature for 16 hours and the thermoelectric power was remeasured at liquid nitrogen temperature without any intervening plastic or elastic deformation.

The imperfection structure partially removed by this mild heat treatment is similar in its effects to the one observed in the experiments of Molenaar and Aarts<sup>2</sup> where electrical resistivity was found to change in a dis-

continuous manner after anneal, the effect being attributed to vacancies.<sup>3</sup> To account for the abrupt changes in value of the thermoelectric power in the intervals preceding point "A" and following (not shown) it is necessary to assume further that large-scale removal of vacancies from their positions in the lattice is possible after the density of population reaches some limit. This removal could be accomplished by accumulation at grain boundaries, escape to the surface of the specimen, or annihilation within the lattice. A similar suggestion was earlier advanced to explain abrupt changes in the ratio of resistance changes to elastic strain,  $(\Delta R/R)/(\Delta L/L)$ , observed in cold-worked copper.<sup>4</sup> These resistance changes are now known to occur simultaneously with a discontinuous change in the thermoelectric power.

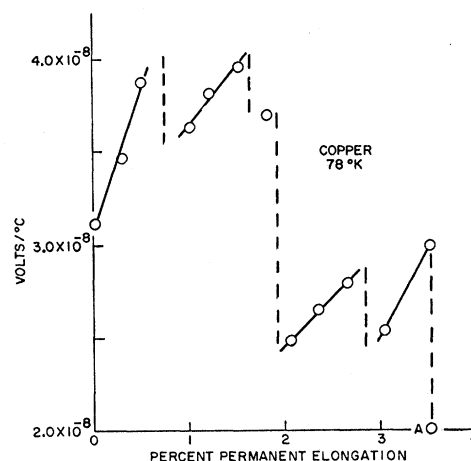


FIG. 1. Variations produced in the thermoelectric power of copper by plastic elongation at liquid nitrogen temperature.

A general trend, having a negative slope in Fig. 1, is superimposed upon the "saw tooth" variation. After sufficient elongation (about 11% in this instance) the points cross the zero axis and the polarity of the couple is reversed, signifying that the structural damage in the specimen portion of the circuit is equivalent to or greater than that present in the reference copper. This trend has been shown by Druyvesteyn and Ooijen<sup>5</sup> to depend upon the increasing dislocation density and can be influenced only by annealing above the recrystallization temperature. If these imperfection structures introduced by cold work have been correctly identified, then the trends shown in Fig. 1 require that the effect of vacancies on the thermoelectric power is of opposite sign to that for dislocations.

<sup>1</sup> C. Crussard, *Bristol Conference on Strength of Solids, 1947* (The Physical Society, London, 1948), p. 119.

<sup>2</sup> J. Molenaar and W. H. Aarts, *Nature* 166, 690 (1950).

<sup>3</sup> T. Broom, *Advances in Phys.* 3, 74 (1954).

<sup>4</sup> E. W. Kammer, *Phys. Rev.* 99, 594 (1955).

<sup>5</sup> M. J. Druyvesteyn and D. J. Ooijen, *Appl. Sci. Research* 5, 437 (1955).