

Effect of Order on the Hall Constants of  $\text{Ni}_3\text{Mn}$ <sup>†</sup>

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(Received July 24, 1961)

Hall effect measurements as a function of temperature are reported for ordered and disordered  $\text{Ni}_3\text{Mn}$ . At low temperatures the ordinary Hall coefficient is found to be negative for both the ordered and disordered alloy. It is shown that the apparent positive value of the ordinary Hall coefficient in the disordered alloy at room temperature is due to the near coincidence of the Curie temperature of this alloy with room temperature. This results in a positive contribution to the measured value of the ordinary Hall coefficient from the extraordinary effect, a smaller contribution being present at low temperatures. An approximate correction is made for this contribution, with the result that the true ordinary coefficient in disordered  $\text{Ni}_3\text{Mn}$  is found to be nearly equal to that in the ordered alloy. Furthermore, its absolute magnitude and temperature dependence are much like that of Cu-Ni and pseudonickel alloys. On this basis it is concluded that ordering has much less effect on the band structure of this alloy than was previously assumed.

## INTRODUCTION

THE Hall effect in ferromagnetics has been found to obey a relation of the form<sup>1</sup>

$$(Vt/I) = R_0(\mu_0 H) + R_1 M, \quad (1)$$

where  $V$  is the Hall potential,  $t$  the sample thickness,  $I$  the sample current,  $H$  the magnetic field, and  $M$  the magnetization. Pugh<sup>1</sup> has designated  $R_0$  and  $R_1$  as the ordinary and extraordinary Hall coefficients, respectively. For a metal having a single conduction band that is either nearly empty or nearly full, the ordinary coefficient  $R_0$  is related to the number of carriers by the equation

$$R_0 = -(1/Ne), \quad (2)$$

where  $N$  is the number of atoms per  $\text{m}^3$ ,  $n$  the number of carriers per atom, and  $e$  the magnitude of the electronic charge ( $1.6 \times 10^{-19}$  coul). With the conventional choice of signs for the directions of the field and current, a negative sign for  $R_0$  indicates electronic conduction while a positive sign indicates conduction by holes. For a multi-band metal, the expression for  $R_0$  is considerably more complicated<sup>2</sup> than that given by Eq. (2); nevertheless, the sign of  $R_0$  is still useful as indicating the sign of the predominant carriers.

In the case of  $\text{Ni}_3\text{Mn}$  it has been known for some time<sup>3</sup> that the measurements seem to indicate a strong dependence of  $R_0$  on the degree of order, varying (at room temperature) from about  $-16 \times 10^{-11} \text{ m}^3/\text{coul}$  for a well ordered sample to about  $+6 \times 10^{-11} \text{ m}^3/\text{coul}$  for a disordered sample. This is quite in contrast with FeCo, where  $R_0$  was found<sup>4</sup> to vary little with order, and has led<sup>5,6</sup> to some very interesting speculations regarding the effect of order on the band structure of  $\text{Ni}_3\text{Mn}$ .

<sup>†</sup> This research was supported by the U. S. Air Force through the Air Force Office of Scientific Research.

<sup>1</sup> E. M. Pugh, N. Rostoker, and A. Schindler, *Phys. Rev.* **80**, 688 (1950).

<sup>2</sup> E. M. Pugh, *Phys. Rev.* **97**, 647 (1955).

<sup>3</sup> E. M. Pugh, *American Institute of Physics Handbook* (McGraw-Hill Book Company, Inc., New York, 1957), Sec. 5, pp. 237-239.

<sup>4</sup> S. Foner, F. E. Allison, and E. M. Pugh, *Phys. Rev.* **109**, 1129 (1958).

<sup>5</sup> J. E. Goldman, *Revs. Modern Phys.* **25**, 108 (1953).

<sup>6</sup> J. B. Goodenough, *Phys. Rev.* **120**, 67 (1960).

It has already been pointed out by Pugh<sup>4</sup> however, that the apparent positive room temperature value of  $R_0$  for a disordered sample of  $\text{Ni}_3\text{Mn}$  does not necessarily indicate conduction by holes, but is rather a reflection of the interesting magnetic properties of this alloy. Generally, the ordinary Hall coefficient is obtained from the high-field slope of a curve of the Hall field per unit current density as a function of the magnetic induction  $B$ . As Eq. (1) shows, this slope is given by

$$R_0^* = (\partial/\partial B)(Vt/I) = R_0 + (R_1 - R_0)\partial M/\partial B. \quad (3)$$

If the sample is truly saturated, then the second term on the right vanishes and  $R_0^*$  reduces to  $R_0$ . For disordered  $\text{Ni}_3\text{Mn}$ , however, the Curie temperature is near room temperature, so that the second term is by no means negligible, even at very high fields. Since  $R_1$  is positive and much greater than  $R_0$  for this alloy, it is then probable that the positive slope is due to the second term having much larger magnitude than the first and that  $R_0$ , in contrast to  $R_0^*$ , is negative. Thus, it is very probable that conduction in disordered  $\text{Ni}_3\text{Mn}$  is by electrons, rather than by holes as has been assumed.<sup>5</sup> This conclusion would also be more in keeping with the results of measurements on the FeCo alloy, where it was found that ordering had little effect on  $R_0$  and consequently, on the band structure. To further verify these conclusions we have extended the Hall effect measurements on an ordered and a disordered sample of  $\text{Ni}_3\text{Mn}$  to lower temperatures, where the second term on the right of Eq. (3) should become small. Actually, this term is still quite large for the disordered sample even at the lowest temperatures and the highest fields. Nevertheless, the results show conclusively that conduction in disordered  $\text{Ni}_3\text{Mn}$  is, as in ordered  $\text{Ni}_3\text{Mn}$ , predominantly by electrons rather than by holes.

## EXPERIMENTAL METHOD AND RESULTS

The samples used for this work were cut from a forged bar furnished by the Westinghouse Manufacturing Company. They were machined into flat plates approximately 5 cm long, 2 cm wide, and 1 mm thick. The

TABLE I. Hall constants and resistivity of  $\text{Ni}_3\text{Mn}$ .

Sample	$T$ (°K)	$R_0^* \times 10^{11}$ (m <sup>3</sup> /coul)	$R_0 \times 10^{11}$ (m <sup>3</sup> /coul)	$R_1 \times 10^{11}$ (m <sup>3</sup> /coul)	$\rho \times 10^8$ (ohm-m)
Ordered	20		-24.0	88	29.1
	77		-22.8	97	30.2
	112		-19.5	126	31.4
	169		-17.8	170	33.9
	302		-12.6	285	40.9
Disordered	20	-14.2	-17	93	66.4
	77	-10.1	-17	240	67.9
	112	-6.5	-17	380	69.2
	169	-6.7	-18	700	70.8
	231	3.4	-13	1100	
	302	9.0			73.9
	322	3.7			

samples were heated to 600°C in a vacuum and cooled rapidly to room temperature to disorder the material. One sample was used as such. The other was annealed for seven days at 470°C, slowly cooled for a period of five days to 350°C, and then cooled to room temperature in one day. Measurements on the latter sample gave a value of 0.893 weber/m<sup>2</sup> for the saturation magnetization at room temperature.

The Hall effect and resistivity of the two samples were measured at a number of temperatures between 20° and 330°K, by a technique previously described.<sup>7</sup> The Hall electric field per unit current density as a function of the magnetic induction is shown for the well-ordered sample in Fig. 1. The figure shows that at each temperature the data give a clearly defined straight line at the higher fields. In each case  $R_0$  is negative and  $R_1$  positive. The results for the disordered sample are shown in Fig. 2. Here, most notably at the higher temperatures, the data have not yet become linear, but the high-field slope appears to change sign somewhere between 112° and 231°K, in contrast with the ordered sample.

The results of the Hall effect measurements shown in Figs. 1 and 2 are listed in Table I, together with the measured values of the resistivity. For the ordered

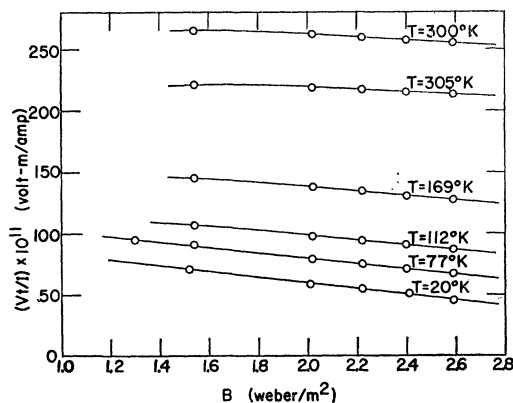


FIG. 1. Hall electric field per unit current density plotted against magnetic induction for ordered  $\text{Ni}_3\text{Mn}$ .

<sup>7</sup> S. Foner and E. M. Pugh, Phys. Rev. **91**, 20 (1953).

sample, the ordinary coefficient  $R_0$  and the extraordinary coefficient  $R_1$  were obtained by fitting the high-field points to the straight line

$$(Vt/I) = R_0 B + (R_1 - R_0) M_s, \quad (4)$$

where  $M_s$  is the saturation magnetization of the sample. For the disordered sample, the slopes of the curves shown in Fig. 2 were estimated in the region of 2.6 webers/m<sup>2</sup>. These slopes are listed in the table as  $R_0^*$ , rather than as  $R_0$ , reflecting the fact that the second term on the right in Eq. (3) is not negligible for the disordered alloy.

The values of  $R_0^*$ , which for the ordered alloy equals  $R_0$ , are plotted in Fig. 3 against the reduced temperature  $T/T_c$ . For the Curie temperatures  $T_c$  we have used the values<sup>8,9</sup> 744° and 270°K for the ordered and disordered alloy, respectively.

## DISCUSSION

The data, as plotted in Fig. 3, show very clearly that the apparent positive value for the ordinary Hall coefficient in disordered  $\text{Ni}_3\text{Mn}$  at room temperature is due to the near coincidence of the Curie temperature of this alloy with room temperature. The curve shown in Fig. 3 should be compared with similar curves by Allison and Pugh<sup>10</sup> for some alloys of Cu in Ni. For those alloys, both  $R_0$  and  $(R_1 - R_0)$  were negative, so that near the Curie temperature the contribution from the second term on the right of Eq. (3) increased the magnitude of the apparent value of  $R_0$  but did not change its sign. For disordered  $\text{Ni}_3\text{Mn}$ , however,  $R_0$  is negative (as shown by the low-temperature data) but  $(R_1 - R_0)$  is positive. Thus the effect here is to change the sign of the apparent value of  $R_0$ .

Since some magnetic data for disordered  $\text{Ni}_3\text{Mn}$  are available in the literature, it is possible to estimate the

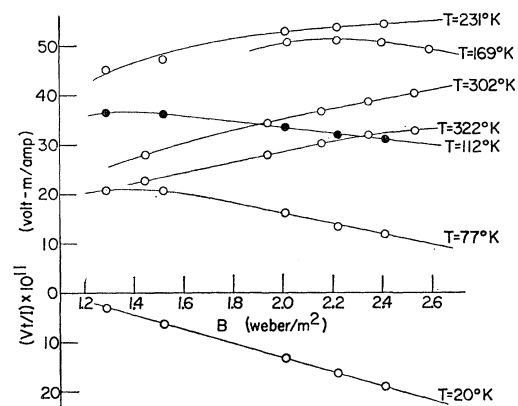


FIG. 2. Hall electric field per unit current density plotted against magnetic induction for disordered  $\text{Ni}_3\text{Mn}$ .

<sup>8</sup> S. Kaya and M. Nakayama, Proc. Phys.-Math. Soc. Japan **22**, 126 (1940).

<sup>9</sup> S. Kaya and A. Kussman, Z. Physik **72**, 293 (1931).

<sup>10</sup> F. E. Allison and E. M. Pugh, Phys. Rev. **102**, 1281 (1956).

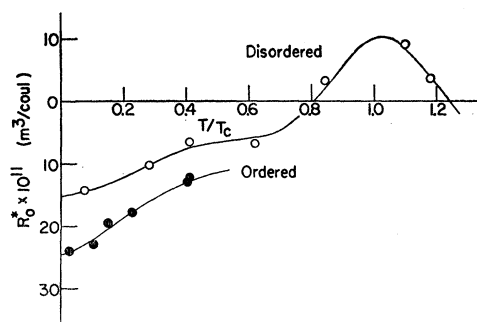


FIG. 3. Observed values of  $R_0^*$  vs  $T/T_c$  for ordered and disordered  $\text{Ni}_3\text{Mn}$ , showing the anomaly near the Curie point.

necessary correction to  $R_0^*$  in order to obtain  $R_0$ . Rather than work with Eq. (3), however, we have used Eq. (1) directly, with values of  $\mu_0 H$  and  $M$  estimated from the work of Kouvel, Graham, and Becker.<sup>11</sup> The values of  $R_0$  and  $R_1$  so obtained are listed for the disordered alloy in Table I. These should be considered as only rough estimates. It is apparent, however, that the correction is very large, even well below the Curie temperature.

The corrected values for the ordinary Hall coefficient in the disordered alloy are not too different from the measured values for the ordered alloy. Furthermore, the plot in Fig. 3 of  $R_0^*$  vs  $T/T_c$  shows that, for  $\text{Ni}_3\text{Mn}$ , the absolute value of  $R_0$  and its temperature dependence are both close to those observed in Cu-Ni alloys<sup>10</sup> and in the ternary alloys of Cu-Ni-Fe that have 28 electrons per atom.<sup>12</sup> On this basis it can be concluded that ordering has much less effect on the band structure of  $\text{Ni}_3\text{Mn}$  than had been assumed.

Various theories<sup>13-15</sup> have recently been proposed regarding the extraordinary Hall coefficient  $R_1$ . These

<sup>11</sup> J. S. Kouvel, C. D. Graham, and J. J. Becker, J. Appl. Phys. **29**, 518 (1958).

<sup>12</sup> E. R. Sanford, A. C. Ehrlich, and E. M. Pugh, Phys. Rev. **123**, 1947 (1961).

<sup>13</sup> R. Karplus and J. M. Luttinger, Phys. Rev. **95**, 1154 (1954).

<sup>14</sup> J. Smit, Physica **21**, 877 (1955).

<sup>15</sup> J. M. Luttinger, Phys. Rev. **112**, 739 (1958).

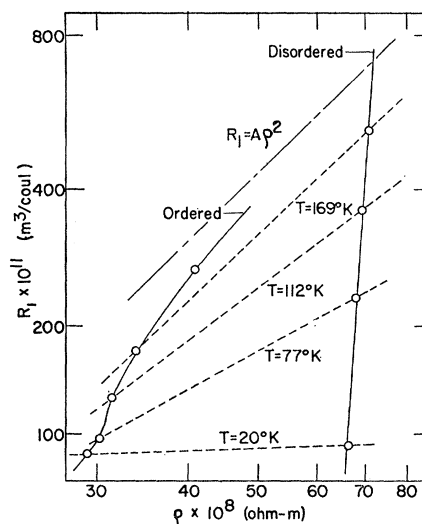


FIG. 4. Extraordinary Hall coefficient for ordered and disordered  $\text{Ni}_3\text{Mn}$  plotted against the resistivity on a log log scale.

theories usually relate  $R_1$  with the resistivity  $\rho$ , and it is customary to plot the extraordinary coefficient as a function of the resistivity on a  $\log R_1$  vs  $\log \rho$  plot. Figure 4 shows such a plot for the data reported here. For comparison, we have included in this figure the line

$$R_1 = A\rho^2.$$

It will be noted that the dashed line connecting the measurements at 169°K is very nearly exactly parallel to this line, as was a similar plot by Foner, Allison, and Pugh<sup>4</sup> of room temperature values for different states of order. Below 169°K, however, this relation becomes an increasingly poorer representation of the data.

#### ACKNOWLEDGMENTS

This study was proposed by Professor Emerson M. Pugh and his suggestions for carrying it out were very helpful. Dr. Floyd E. Allison also contributed useful suggestions and performed many of the measurements.