Thermoremanent magnetization and blocking temperatures in Ni_3Mn

R. B. Goldfarb and C. E. Patton

Department of Physics, Colorado State University, Fort Collins, Colorado 80523

Initial susceptibility and thermoremanent magnetization measurements have been made on atomically disordered and weakly ordered Ni_3Mn. The blocking of superparamagnetic clusters yields a peak in initial susceptibility near 110 K. Ferromagnetic clusters with Curie temperatures near 265 K cause a break in the susceptibility vs. temperature curves at 265 K. Exchange anisotropy effects appear below 35 K.

PACS numbers: 75.30.Cr, 75.30.Kz, 75.20.En, 75.60.Nt

INTRODUCTION

Atomically disordered and weakly ordered nickel-manganese alloys near the Ni_3Mn composition have magnetic properties which can phenomenologically be described as mictomagnetic.[1] One of these properties is a unidirectional anisotropy induced by field cooling to liquid helium temperatures, which causes a shifted hysteresis loop and a thermoremanent magnetization (TRM) in the direction of the cooling field.[2] Such displaced hysteresis loops are evidence of exchange anisotropy, a term first applied to the interaction between ferromagnetic Co and antiferromagnetic CoO.[3] In disordered Ni-Mn and related alloys, the concept has been applied to ferromagnetic and antiferromagnetic domains[4] as well as interactions at the nearest-neighbor atomic level.[5]

Another characteristic of mictomagnets is a peak in initial susceptibility vs. temperature. In dilute spin glasses such peaks can be rather sharp, with a width of only a few degrees. In mictomagnets, including Ni-Mn alloys, the susceptibility peaks are rather broad. Wohlfarth has recently pointed out that such peaks are often seen in rock magnetism.[6] Furthermore, TRM has long been observed in magnetic rocks cooled from high temperature in the earth's field and was attributed by Néel to the blocking of essentially superparamagnetic grains.[7]

The present paper describes measurements of initial susceptibility and thermoremanent magnetization in atomically disordered and weakly ordered Ni_3Mn. The data indicate that the peak in susceptibility is due to the blocking of superparamagnetic clusters. Exchange anisotropy effects appear below 35 K, much below the susceptibility peak.

EXPERIMENT

The alloy, 24.6 at.% Mn-Ni, was prepared as described in Ref. 5. A disordered sample approximately 3 mm in diam was obtained by homogenizing for 9 hrs at 1000°C and quenching. Weak atomic order was obtained by annealing an initially disordered sample in evacuated Pyrex capsules at 500°C which were then shattered in ice water by a mouse-trap device. Annealing times were taken to be cumulative.

Susceptibility measurements were made by a low-field ac method. A primary field coil was wound on a coilform made of glass fiber-filled phenolic, with a coefficient of thermal expansion of $1 - 2 \times 10^{-5}$ cm/cm-°C. Two secondary coils were wound in opposition over the primary coil, one to measure the emf induced by the sample and one to compensate for the emf induced by the primary coil. The sample for measurement was located within the coilform and the measuring secondary. A carbon resistor temperature sensor[8] was located 0.5 cm from the primary coil. The entire magnetometer was surrounded by a thin copper isothermal shield with slits to inhibit eddy currents. The magnetometer assembly, 9 cm long by 1 cm in diam, was fitted to a stainless steel tube and inserted directly inside a liquid helium storage dewar.[9] The sample temperature was controlled by lowering the probe into the dewar by an appropriate distance, taking advantage of the temperature gradient between the top of the dewar and the liquid level. The lowering was done in small steps. The temperature and susceptibility were recorded after the magnetometer achieved thermal equilibrium, every few degrees.

The primary solenoid was driven with a 15 mA rms constant current source at 20 Hz, giving an rms field of 2.5 Oe. Additionally, one set of measurements was made as a function of frequency from 2 to 20,000 Hz. Using the magnetic dipole moment of a spherical sample and Faraday's law, the absolute external susceptibility was deduced from the net emf induced on the secondary. The internal susceptibility was calculated by correcting for sample demagnetization. These values agree to within 10% with low-field dc measurements.

Superparamagnetic cluster moments were determined from a Langevin superparamagnetism model by the method described in Ref. 10 in the temperature regime above the susceptibility peak. Dc magnetization measurements were made with a vibrating-sample magnetometer as a function of cooling field and temperature. The samples, initially demagnetized, were cooled from 300 to 4 K in dc fields of 50 to 8800 Oe. Thermoremanent magnetization at 4 K was measured with the electromagnet removed from the vicinity of the sample. A zero-field-cooled sample was measured in 8000 Oe from 4 to 300 K.

RESULTS AND DISCUSSION

The results of the ac susceptibility measurements are shown in Figs. 1 to 4. Figure 1 shows normalized internal susceptibility as a function of temperature for the disordered sample and two weakly ordered samples. The relatively wide peaks can be attributed to a distribution in blocking temperatures due to a fluctuation in local composition of the magnetic clusters and a distribution in cluster sizes. The susceptibility peaks broaden and move to higher temperature as ordering occurs. The peak susceptibility χ_B and temperature T_B are shown in Fig. 2 as a function of annealing time at 500°C. The peak shift to higher temperatures is due to the increase in size of the magnetic clusters with ordering, as shown in Ref. 11. The overall increase in susceptibility is due to an increase in cluster moment.

 0021-8979/79/117358-03$01.10 © 1979 American Institute of Physics

Downloaded 09 May 2003 to 129.82.31.230. Redistribution subject to AIP license or copyright, see http://ojps.aip.org/japo/japcr.jsp

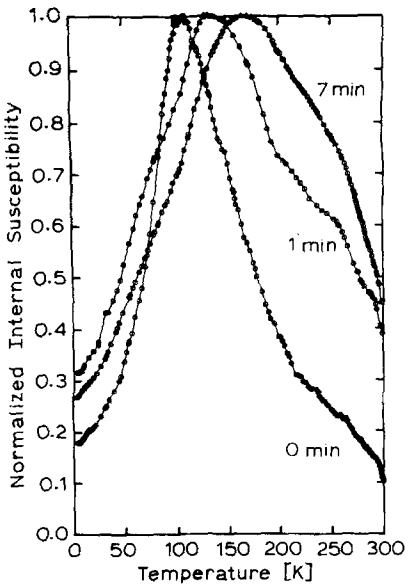

Fig. 1. Normalized internal susceptibility vs. temperature for disordered and weakly ordered 24.6 at.% Mn-Ni. Annealing times at 500°C for the initially disordered alloy are indicated.

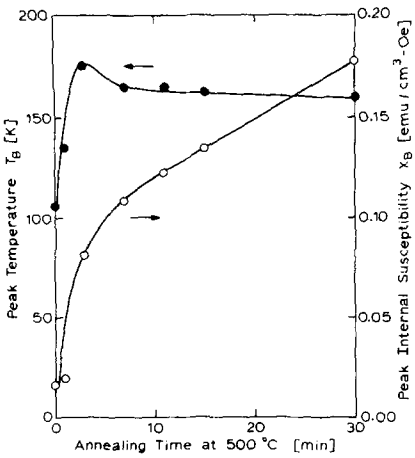

Fig. 2. Peak internal susceptibility and peak temperature as a function of annealing time at 500°C.

A set of measurements was performed on another disordered Ni-Mn sample with slightly different T_B. In this experiment, the measurement frequency was varied from 2 to 20,000 Hz. Figure 3 shows an Arrhenius plot of inverse frequency vs. inverse T_B. The linear variation is consistent with a blocking model and the slope yields a thermal activation energy of 1.6 eV. This linear variation also provides evidence that the peak is not connected with antiferromagnetic ordering or an increase in crystalline anisotropy at low temperature.

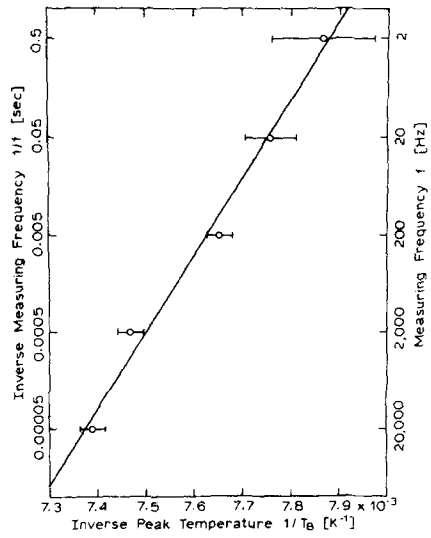

Fig. 3. Arrhenius plot of inverse measuring frequency vs. inverse susceptibility peak temperature for a disordered 24.6 at.% Mn-Ni sample.

The inflection point in χ vs. T for the disordered sample at 265 K and notable fall-off in χ above 265 K for the weakly ordered samples indicate the presence of ferromagnetic clusters with a Curie point at about this temperature. The same 265 K Curie temperature is also evident in the external susceptibility data for ferromagnetic, atomically disordered 22.1 at.% Mn-Ni in Fig. 4. This Curie temperature is quite different from the 725 K Curie temperature of fully ordered Ni_3Mn.

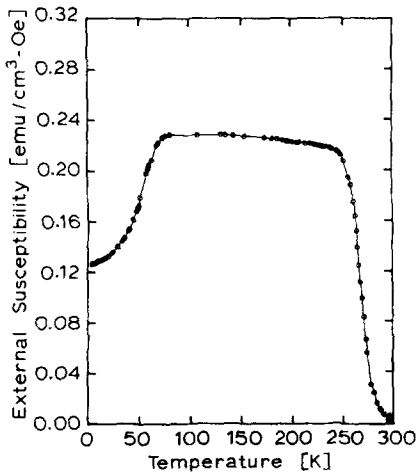

Fig. 4. External susceptibility vs. temperature for a disordered 22.1 at.% Mn-Ni spherical sample. Below 265 K, external susceptibility approaches the reciprocal of the demagnetizing factor as expected for a soft ferromagnet.

Downloaded 09 May 2003 to 129.82.31.230. Redistribution subject to AIP license or copyright, see http://ojps.aip.org/japo/japcr.jsp

Now consider the dc magnetization data. Figure 5 shows the magnetization of disordered 24.6 at.% Mn-Ni upon warming in an external dc field of 8000 Oe after zero-field cooling from 300 to 4 K. It is noteworthy that the peak magnetization occurs at 35 K, significantly below the susceptibility peak temperature of 110 K. This peak at 35 K is not simply a shifted susceptibility peak, related to the data in Fig. 1. A Langevin analysis of magnetization vs. applied field for the same sample from above T_B to 300 K indicates a superparamagnetic cluster moment of about 600 μ_B. Such clusters would be almost completely field-aligned in a field of 8000 Oe below 35 K; any blocking effect would be suppressed. The magnetization peak at 35 K, therefore, is not connected with the blocking of superparamagnetic clusters that dominates the susceptibility results discussed above. Rather, it can be attributed to the decay of exchange anisotropy effects upon warming. Reversible hysteresis loops, originally shifted after field cooling to liquid helium temperatures, shift back to the origin upon warming to 35 K.[2] Indeed, it was shown that measuring fields of 50,000 Oe are necessary to override the drop in magnetization below 35 K.[5]

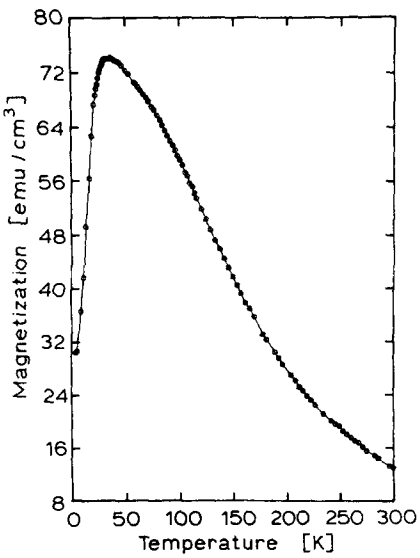

Fig. 5. Magnetization in 8000 Oe field as a function of increasing temperature after zero-field cooling for the same disordered 24.6 at.% Mn-Ni sample as in Fig. 1.

Figure 6 shows the TRM obtained by field cooling to 4 K as a function of cooling field for a single 24.6 at.% Mn-Ni sample in the atomically disordered state and for weak order after annealing at 500°C for the times indicated. The data for the disordered sample, labeled "0 min", show a simple monotonic increase in the TRM with cooling field. This is in agreement with previous results.[2] With weak ordering, however, the shape of the TRM-cooling field curve changes radically. Figure 6 shows that the TRM increases to a maximum for a cooling field of about 600 Oe and then decreases. In addition, the TRM at 8800 Oe initially decreases as ordering is initiated and then increases for annealing times in excess of about 3 minutes.

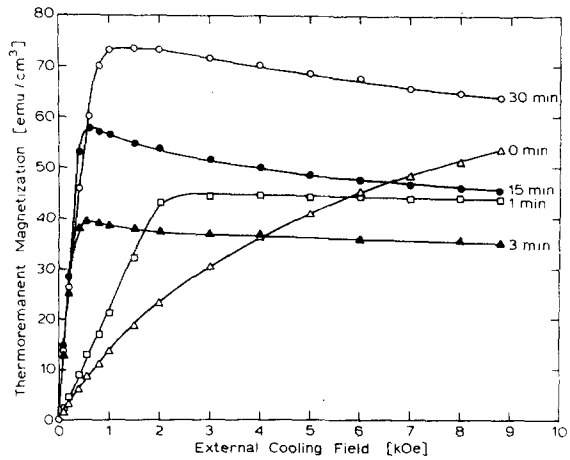

Fig. 6. Thermoremanent magnetization at 4 K as a function of cooling field for a 24.6 at.% Mn-Ni sample. The cumulative annealing times at 500°C for the initially disordered alloy are indicated.

The decrease in the TRM for cooling fields greater than 600 Oe is evidence for "hard" magnetic moments, presumably Mn moments, aligned roughly antiparallel to the cooling field. A model based on such antiparallel Mn moments is discussed in detail in Ref. 12, in connection with a peculiar remanence reversal effect in these alloys. It is apparent that annealing at 500°C produces, in addition to the nucleation and growth of "soft" ferromagnetic clusters with Curie temperatures of 265 K, a population of "hard" moments antiparallel to the net magnetization which tend to reduce the TRM.

ACKNOWLEDGEMENTS

The authors thank A. B. Tveten for helpful suggestions and discussions. Parts of this work were supported by the National Science Foundation, Grant No. DMR76-23623.

REFERENCES

1. P. A. Beck, Prog. Mater. Sci. 23, 1 (1978).
2. J. S. Kouvel and C. D. Graham, J. Phys. Chem. Solids 11, 220 (1959).
3. W. H. Meiklejohn and C. P. Bean, Phys. Rev. 102, 1413 (1956).
4. J. S. Kouvel, J. Phys. Chem. Solids 24, 795 (1963).
5. T. Satoh, R. B. Goldfarb and C. E. Patton, J. Appl. Phys. 49, 3439 (1978).
6. E. P. Wohlfarth, Physica 86-88B, 852 (1977).
7. L. Néel, Adv. Phys. 4, 191 (1955).
8. J. R. Clement and E. H. Quinnell, Rev. Sci. Inst. 23, 213 (1952).
9. It was important to use a dewar with a wide neck to avoid induction from the walls. "Cryenco" brand was satisfactory.
10. A. A. v.d. Giessen, J. Phys. Chem. Solids 28, 343 (1967).
11. N. V. Volkenshtein, N. I. Kourov and Yu. N. Tsiovkin, Fiz. Metal. Metalloved. 42, 213 (1976).
12. T. Satoh, R. B. Goldfarb and C. E. Patton, Phys. Rev. B 18, 3684 (1978).

Downloaded 09 May 2003 to 129.82.31.230. Redistribution subject to AIP license or copyright, see http://ojps.aip.org/japo/japcr.jsp