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# SHORT RANGE ORDER DEVELOPMENT IN $Ni_3Mn$ ALLOYS*

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## ABSTRACT

The magnetic order development in polycrystalline $Ni_3Mn$ alloys has been studied as a function of atomic ordering during anneal at 500°C for times up to 32 hours after an initial quench from 1000°C. The cluster moments appear to saturate at about $5 \times 10^{-17}$ emu, with an exponential growth characterized by a time constant of about 5-10 hours. The clusters grow to about 40 Å radius, assuming an aligned moment of 1 $\mu_B$ per atom within clusters. The cluster density increases abruptly to $2.0 \times 10^{19}$ $cm^{-3}$ after the initial 5-minute anneal, levels off, and then decays to about $1.0 \times 10^{19}$ $cm^{-3}$. These data suggest that the clusters "pop in" during the initial stages of short range order development, and then grow in size to saturation.


## I. INTRODUCTION

Nickel manganese alloys near the $Ni_3Mn$ composition show large changes in magnetic properties with the degree of short range order. Kneller[1] originally reported the superparamagnetic character of the alloy after 20 hours anneal at 480°C, with cluster sizes on the order of 30 Å. Following this work, Marcinkowski and Brown[2] studied the short range atomic order development by neutron diffraction, coupled with limited magnetic measurements using ballistic methods at relatively low fields (100 Oe).

The purpose of this study was to characterize the cluster dynamics for superparamagnetic $Ni_3Mn$ alloys during short range order (SRO) development. The method of analysis follows that of Kneller[1]. Data on moment vs. field at room temperature for different annealing times at 500°C, following an initial quench from 1000°C to produce a "disordered" alloy, are fitted to a simple Langevin superparamagnetism model to obtain cluster moment and density versus annealing time. The annealing time is roughly proportional to the degree of short range order development, up to short range order saturation at 20-40 hours at 500°C[2]. No attempt was made at this stage to include the effect of a distribution in cluster moments[1], the effect of interactions by a molecular field formalism, or a nonzero susceptibility for the disordered host environment of the clusters. For this reason, the results may be taken as a qualitative, but not quantitative, description of the cluster growth processes during ordering.

## II. RESULTS

Typical magnetization data at room temperature are shown in Fig. 1 for a 25 at % Mn alloy, versus annealing time at 500°C following the initial quench from 1000°C to room temperature. These data show the monotonic increase in moment, along with a very abrupt increase after the initial 5-minute (0.08 hour) anneal. These features are indicative of the increase in net cluster moment with annealing time for SRO domains.

These data were then analyzed to obtain: (1) the extrapolated magnetization at infinite field ($M_\infty$), and (2) the initial susceptibility at zero field ($\chi_{initial}$). These two parameters, obtained for each magnetization curve for samples annealed at 500°C for times up to 32 hours, were used to calculate average cluster moment $\mu$ and cluster density N versus annealing time. For simple Langevin superparamagnetism, the magnetization M vs. internal field H is given by

$$M = N\mu \mathcal{L}(\mu H/kT), \quad (1)$$

where $\mathcal{L}(x)$ denotes the Langevin function

$$\mathcal{L}(x) = \mathrm{Coth}(x) - (1/x), \quad (2)$$

k is Boltzman's constant, and T is the temperature in K. The limits of $x >> 0$ and $x = 0$ yield two simple working equations

$$M_\infty = N\mu, \quad (3)$$

$$\chi_{initial} = N\mu^2/3kT . \quad (4)$$

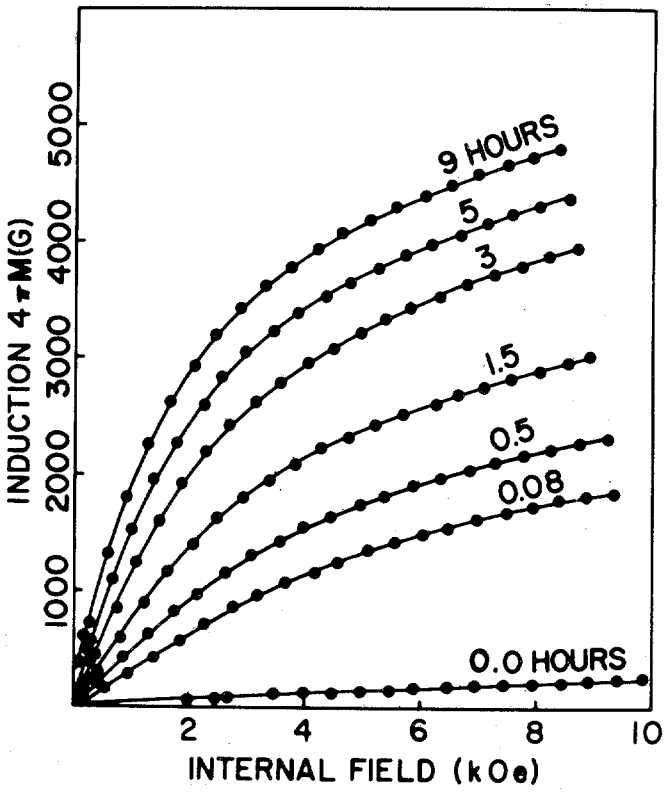


Fig. 1. Magnetic induction $4\pi M$ versus internal field at room temperature with annealing time at 500°C as a parameter, for a 25 at % Mn alloy, following an initial quench from 1000°C to room temperature.

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Results for cluster moment and cluster density versus annealing time are shown in Figs. 2 and 3, respectively. Each figure shows results for three different samples of the same nominal 25 at % Mn composition. The sample-to-sample deviations and the deviation of individual points from the general trend of the data are representative of the error bars on the results. Figure 2 shows the rather rapid increase in cluster moment during the initial stages of SRO development. In the initial 5-minute anneal, the cluster moment doubles, and continues to increase fairly rapidly for 2-3 hours of anneal. At this point, the growth rate slows considerably, but does continue to increase. At twenty hours, the cluster moment is nearly stabilized. Little additional increase is seen during anneal up to 32 hours.

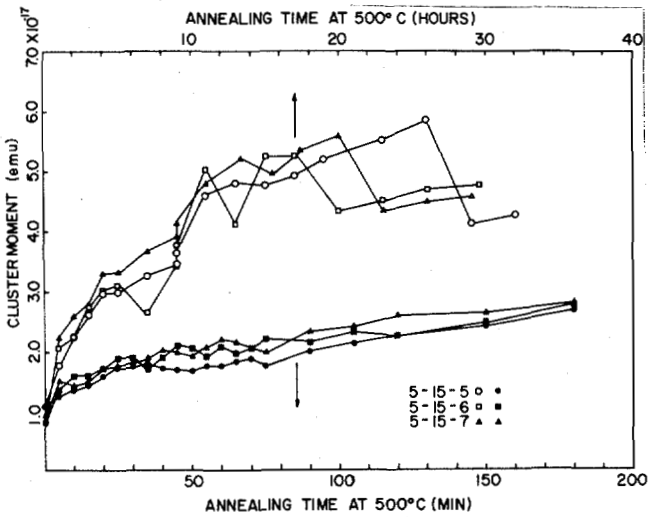


Fig. 2. Calculated cluster moment versus annealing time at 500°C for the three spherical samples of the 25 at % Mn alloy.

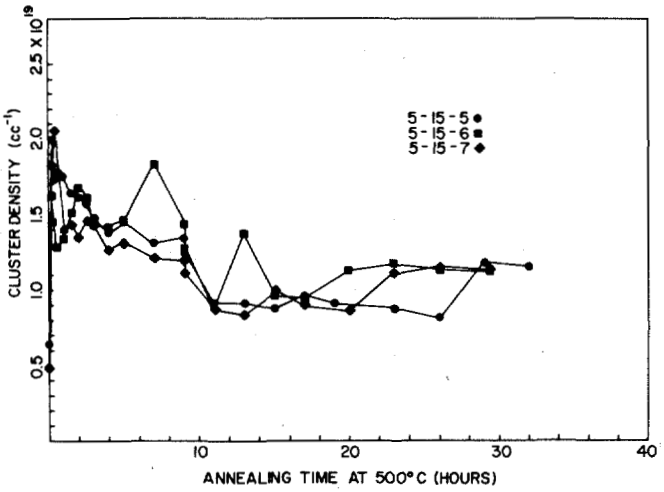


Fig. 3. Calculated cluster density versus annealing time at 500°C for three spherical samples of the 25 at % Mn alloy.

Figure 3 reveals a quite different behavior for the cluster density as a function of annealing time. The density shows a very abrupt increase during the first five minutes of anneal, followed by a period from 5 minutes to 5-10 hours where the cluster density slowly decreases from $2 \times 10^{19}$ to $1 \times 10^{19}$ clusters/cm$^3$. Beyond this time, the density is relatively constant.

## III. CONCLUSIONS

The combined results in Figs. 2 and 3 reveal the qualitative character of short range order related magnetic cluster growth in superparamagnetic $Ni_3Mn$. From an initially "disordered" state, achieved by a rapid oil quench from 1000°C to room temperature, the short range order domains appear to "pop in" during the initial 5-minute annealing treatment at 500°C. Once these initial clusters have nucleated, with a density on the order of $1.5 - 2.0 \times 10^{19}$ per cm$^3$, no additional clusters form with increased heat treatment. Those already formed do continue to grow rapidly for the first 3-5 hours of anneal. After 3-5 hours, the growth continues to saturation at 20 hours. As the growth process occurs, the cluster size becomes sufficient for some clusters to coalesce. Such cluster merging is evidenced by the decrease in the cluster density.

## REFERENCES

1. E. Kneller, "Temperaturabhängigkeit der spontanen Magnetisierungin kleinen ferromagnetischen Teilchen," Z. Physik 1958, Vol. 152, p. 574.

2. M. J. Marcinkowski and N. Brown, "Transformation-Disorder to Order in $Ni_3Mn$," J. Appl. Phys. 1961, Vol. 32, p. 375.