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The effect of pressure on the Curie temperature in Fe–Ni Invar mechanical alloys

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Abstract
Measurements of the temperature dependence of the AC susceptibility were made for Fe–Ni Invar mechanical alloys under hydrostatic pressures up to 1.5 GPa. The Curie temperatures decreased linearly with pressure. The rate of decrease became larger for specimens annealed at higher temperatures. The temperature of annealing after ball milling has been directly related to the extent of the chemical concentration fluctuation, and the extent becomes smaller for specimens annealed at higher temperature. This tendency can be explained by assuming a Gaussian distribution function.

1. Introduction

Fe–Ni alloys around the Invar composition of 35 at.% Ni show various anomalies in both mechanical and magnetic properties [1, 2]. A great number of experimental and theoretical studies on this alloy have accumulated. Anomalies in physical properties such as low thermal expansion coefficient were interpreted as being due to the large positive value of the magnetovolume effect which cancels the normal part of the thermal expansion caused by the anharmonic terms in the lattice vibration.

The anomalies in magnetic properties seen in Fe–Ni Invar alloys are the unusual decrease of the spontaneous magnetization with increasing Fe concentration and the increase of the high-field susceptibility. These anomalies have been understood as a result of the instability of the 3d-band ferromagnetism in the fcc lattice [3]. Due to the existence of a large sharp peak at the top of the 3d-electron band of the fcc phase, the ferromagnetic state becomes energetically

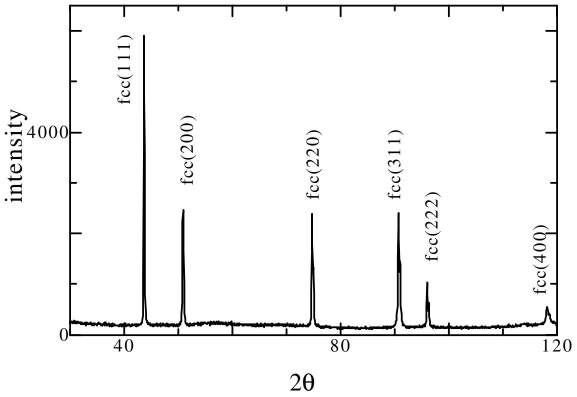

Figure 1. The x-ray pattern observed for Fe–32.4 at.% Ni Invar alloy annealed at 800 °C after mechanical alloying.

unstable when the number of outer electrons is decreased by decreasing the Ni concentration beyond the Invar region.

According to the band picture [3], the ferromagnetic state becomes unstable at a critical concentration of Ni, and therefore, the spontaneous magnetization decreases abruptly at this point. However, the observed anomalous decrease of the saturation magnetization from the Slater–Pauling curve is not so sharp as expected from the model of 3d-band ferromagnetism.

To interpret the actual curvature of the decrease of the saturation magnetization from the Slater–Pauling curve, several models have been proposed taking into account the existence of fluctuation of the alloy concentration. Kachi *et al* [4] considered a Gaussian distribution of the concentration fluctuation and explained the experimental curve.

By mechanical alloying and subsequent annealing, it was possible to control the degree of concentration fluctuation in Fe–Ni Invar alloys [5–8]. The interrelationship between the extent of the concentration fluctuation and the annealing temperature was established [8] by comparing observed Curie temperatures for mechanically alloyed and annealed specimens with calculated ones, by multiplying the Curie temperature versus Ni concentration curve for bulk materials by a Gaussian function.

It seems of interest to investigate the effect of pressure on the Curie temperature in alloys having a wide variety of concentration fluctuations.

2. Experiment

Powders of the original 99.9% pure metal elements Fe and Ni were milled for 75 h in a planetary ball mill in an argon atmosphere using stainless steel balls and containers. The products were annealed at various temperatures between 400 and 1000 °C for 1 h in shielded vacuums and then quenched to room temperature. The average concentration of the specimens in the present experiments was 32.4 at.% Ni. It was shown that the width of the concentration fluctuation is least for the sample annealed at the highest temperature, and becomes larger for samples annealed at lower temperatures.

The products were examined by means of x-ray diffraction, and proved to consist of fcc phase. The observed x-ray patterns for mechanical alloys annealed at 800 and 1000 °C are shown in figures 1 and 2, respectively.

AC susceptibility measurements were made under hydrostatic pressures up to 1.4 GPa by using a gas-pressure-operated high-pressure cell made of nonmagnetic Cu–Be alloy.

The maximum temperature for the present measurements was limited to just above 400 K due to the plastic materials adopted for the high-pressure cell.

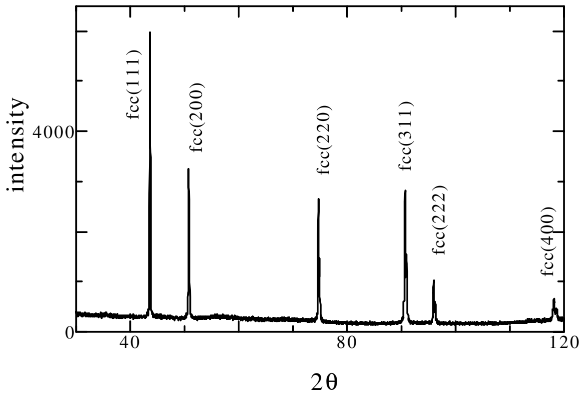

Figure 2. The x-ray pattern for Fe–32.4 at.% Ni mechanical alloy annealed at 1000 °C.

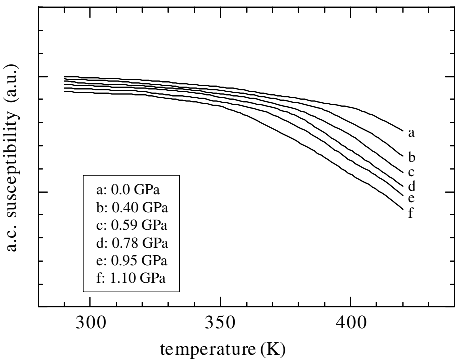

Figure 3. Temperature dependences of the AC susceptibility under various pressures in Fe–32.4 at.% Ni alloy annealed at 800 °C after mechanical alloying.

3. Results and discussion

Observed susceptibility–temperature curves under various pressures for Fe–32.4 at.% Ni Invar alloy annealed at 800 °C after mechanical alloying are shown in figure 3. As the highest attainable temperature was limited to 400 K, it was difficult to obtain the full susceptibility–temperature curve. However, the curves seen in figure 3 are almost parallel to each other, and it is possible to obtain a relative change in T_C from the initial decrease of the start points of those curves. Observed susceptibility versus temperature curves under various pressures for the mechanical Invar alloy annealed at 1000 °C are shown in figure 4. In this figure it is seen that T_C also decreases with pressure. It was not possible to observe AC susceptibility–temperature curves for mechanical alloys annealed below 600 °C, as those alloys have higher Curie temperatures above the limit of the present experimental apparatus.

The Curie temperatures thus determined for mechanical Invar alloys annealed at 800 and 1000 °C are plotted in figure 5 against pressure. In this figure it is seen that the rate of decrease in 1000 °C annealed specimen is larger than that of the 800 °C annealed one which has a wider concentration fluctuation.

This tendency can be explained by considering the effect of the concentration fluctuation as follows. It is known that the pressure coefficient of the Curie temperature in Fe–Ni alloys increases anomalously with decreasing Ni concentration. The average concentration of the present specimen is almost at the peak. Therefore, when the concentration fluctuation becomes wider the average Curie temperature decreases due to the smaller contribution from both sides of the peak.

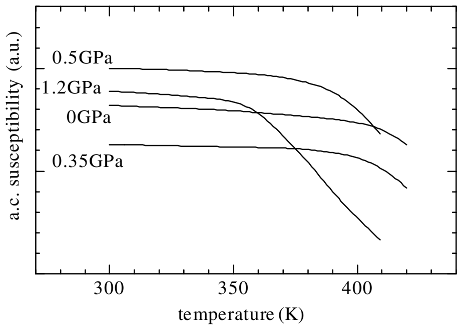

Figure 4. AC susceptibility–temperature curves under various pressures for Fe–32.4 at.% Ni alloy annealed at 1000 °C after mechanical alloying.

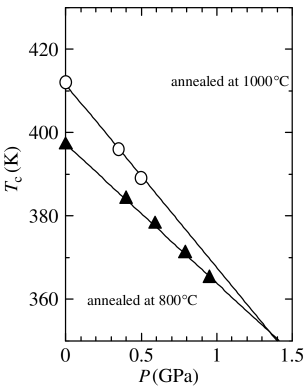

Figure 5. Pressure dependences of the Curie temperature for mechanically alloyed Fe–Ni Invar alloys annealed at 800 and 1000 °C.

4. Conclusions

Pressure dependences of the Curie temperature for mechanically alloyed Fe–Ni Invar alloys have been investigated up to 1.4 GPa. The Curie temperature decreased linearly with pressure. The rate of decrease became smaller for alloys with wider concentration fluctuations. This fact was explained by assuming a Gaussian distribution function for the concentration fluctuation.

Acknowledgment

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