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The influence of plastic deformation on the magnetic properties of $Ni_{50}Cu_{50}$ alloy

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Abstract. Magnetization is measured in plastically deformed $Ni_{50}Cu_{50}$ alloy in the temperature range from 4.5 to 80 K.The temperature dependence of spontaneous magnetization $M_s(T)$ has been analysed according to the self-consistent renormalization theory of spin fluctuation. The value of $M_s(0)$ decreases from 7.07×10^{-2} to 6.32×10^{-2} Wb m^{-2} and the Curie temperature T_c declines from 57.8 to 50.1 K with 50% strain. The decreases in $M_s(0)$ and T_c can be explained by the introduction of the antiphase boundary, which reveals the existence of an atomically ordered structure in the Ni–Cu alloys.

The nickel–copper alloy system has received considerable attention in the investigation of the ferromagnetism of disordered alloys. As is known, nickel and copper, two neighbouring atoms in the periodic table, have the same crystalline structure and extremely similar atomic sizes and electron affinities. Moreover they form a substitutional solid solution alloy with a face-centred cubic structure over the entire range from the pure noble metal to the pure transition metal. This kind of alloy is the most suitable for observing the variation in the electron energy band with alloying. Over the past few decades, considerable experimental and theoretical effort has been made with respect to Ni–Cu alloys. The magnetic properties of these alloys have been explained by consideration of the electron energy band structure. The research results have successfully formed a reference base for studying other alloys.

Nevertheless, there remain several old and still unsolved important issues with regard to Ni–Cu alloys. One is the existence of an order–disorder transformation (Chevenard 1926). Although the existence of an atomically ordered phase in Ni–Cu has been reported in several studies, no direct indication of an atomic order–disorder change has been found yet, since the difference between the x-ray or electron scattering factors of Ni and Cu atoms is too small. The existence of the ordered phase is still quite controversial. Another issue is the deviation of the spontaneous magnetization from the Slater–Pauling curve at high copper concentrations (Aldred 1966). Although many hypotheses have been proposed to explain this deviation, none of them seem completely satisfactory. The magnetism of alloys is sensitive to the atomic arrangement. Recently, we have found that the spontaneous magnetization $M_s(T)$ and the Curie temperature T_c decrease markedly on plastic deformation of the $Ni_{50}Cu_{50}$ alloy. In this paper we report the experimental results on $M_s(T)$ and T_c in the plastically deformed Ni–Cu alloy and investigate the mechanism of the magnetic changes in the properties. We are also interested in explaining the magnetism of Ni–Cu alloys by the self-consistent renormalization (SCR) theory of spin fluctuation (Moriya and Kawabata 1973) and extending the validity of the SCR theory of spin fluctuations to the plastically deformed alloys.

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The raw materials used for alloying in the present investigation were Ni (99.95 wt.%) and Cu (99.99 wt.%). $Ni_{50}Cu_{50}$ alloy was prepared by arc melting the raw materials four times to attain chemical homogeneity on a water-cooled copper hearth in an argon gas atmosphere at a pressure of approximately 93 kPa. The button was homogenized at 1273 K for 72 h and then cooled to room temperature at the rate of 100 K h^{-1}. Rectangular prisms (5 mm × 2.5 mm × 2.5 mm) were cut from the heat-treated button and were compressed at room temperature. The samples for the magnetic measurement were formed with dimensions of 2.5 mm × 2.5 mm × 2.5 mm. The steady field magnetization was measured with a SQUID magnetic fluxmeter (Quantum Design) in the temperature range from 4.5 to 80 K.

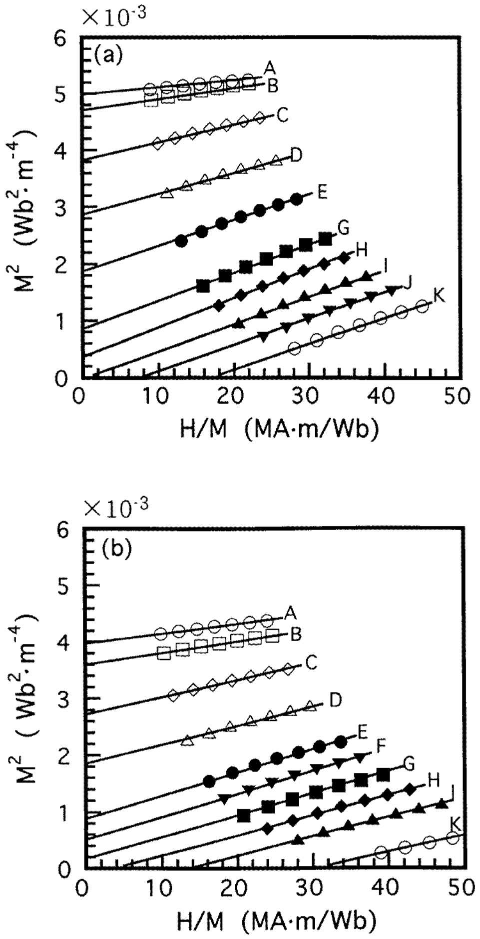

Figure 1. The Arrot plots of the magnetization $M(T, H)$ in $Ni_{50}Cu_{50}$ (a) before plastic deformation and (b) after plastic deformation with 50% strain: curve A, 4.5 K; curve B, 10 K; curve C, 20 K; curve D, 30 K; curve E, 40 K; curve F, 45 K; curve G, 50 K; curve H, 55 K; curve I, 60 K; curve J, 65 K; curve K, 70 K.

Figures 1(a) and 1(b) show the change in the magnetization $M(H, T)$ in the form of the Arrot plots of $[M(H, T)]^2$ versus $H/M(H, T)$ for $Ni_{50}Cu_{50}$ before plastic deformation and after plastic deformation by 50%, respectively. Both can be represented well in the form of $[M(H, T)]^2 = M_s(T) + bH/M(H, T)$ at every temperature, except in the region

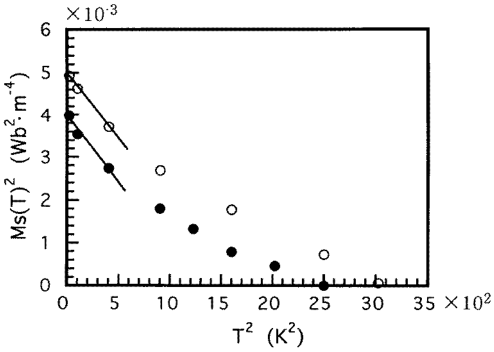

Figure 2. The temperature dependence of the spontaneous magnetization as shown by $[M_s(T)]^2$ against T^2 in $Ni_{50}Cu_{50}$ before plastic deformation (○) and after plastic deformation by 50% strain (●).

of weak magnetic fields. The spontaneous magnetization $M_s(T)$ at T can be obtained by extrapolation of the linear relation to $H = 0$. The plot of $[M(H, T)]^2$ versus $H/M(H, T)$ is commonly used to determine T_c. The values of T_c before and after plastic deformation are 57±2 K and 52±2 K, respectively.

In the SCR theory of spin fluctuation (Moriya and Kawabata 1973), the temperature dependence of $M_s(T)$ has the form $[M_s(T)]^2 = [M_s(0)]^2 - \eta T^2$ at the low temperatures and $[M_{s(}T)]^2 = \xi(T_c^{4/3} - T^{4/3})$ in a fairly wide temperature range below T_c. Figure 2 shows the data of $[M_s(T)]^2$ versus T^2. $[M_s(T)]^2$ shows a sharp decrease at low temperatures and a gradual decrease at higher temperatures with T^2. The spontaneous magnetization $M_s(0)$ at 0 K is determined by extrapolation of this linear relation to $T = 0$ K. The value of $M_s(0)$ without plastic deformation is 7.07×10^{-2} Wb m^{-2}. The average magnetic moment per atom for the sample before plastic deformation is calculated to be $0.069\mu_B$. Here μ_B is the Bohr magneton. Ahern *et al* (1958) measured $M_s(0)$ of Ni–Cu alloys and showed that $M_s(0)$ for $Ni_{51.3}Cu_{48.7}$ is 7.62×10^{-2} Wb m^2. $M_s(0)$ for the undeformed sample in the present study is rather close to their result. The value of $M_s(0)$ for the sample strained by 50% is 6.32×10^{-2} Wb m^2. The plastic deformation produces a significant reduction in $M_s(0)$.

The plots of $[M_s(T)]^2$ against $T^{4/3}$ for the samples before and after plastic deformation are shown in figure 3. $[M_s(T)]^2$ decreases linearly with $T^{4/3}$ over the whole temperature range, indicating that the undeformed and deformed samples are in good consistency with the SCR theory of spin fluctuation. T_c can be obtained by the extrapolation of the linear relation to $M_s(T) = 0$. $T_c = 57.8$ K for the $Ni_{50}Cu_{50}$ alloy without plastic deformation and T_c decreases to 50.1 K on plastic deformation after 50% strain. These values are close to those obtained by the Arrot plots in figure 1. T_c for the undeformed sample is close to that of Ahern *et al* (1958) in which T_c was obtained by plotting $[M_s(T)]^2$ against T for small values of $M_s(T)$ and extrapolating to $[M_s(T)]^2 = 0$.

The average magnetic moment per atom as a function of the number of outer electrons in transition metals and alloys is known as the Slater–Pauling curve (Bozorth 1951). The concentration dependence of the magnetic moment, $\mathrm{d}\mu/\mathrm{d}c$, for Ni-based alloys containing impurities decreases approximately linearly with $-Z\mu_B$ (Marian 1937, Ahern *et al* 1958, Crangle and Marian 1959, Crangel and Parsons 1960, Van Elst *et al* 1962). Here Z is a constant close to the difference between the outer electron numbers of Ni and impurity atoms.

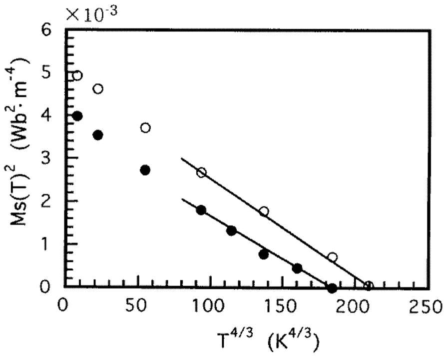

Figure 3. The temperature dependence of the spontaneous magnetization as shown by $[M_s(T)]^2$ against $T^{4/3}$ for $Ni_{50}Cu_{50}$ before plastic deformation (○) and after plastic deformation by 50% strain (●).

Ahern *et al* (1958) measured precisely Z in Ni–Cu alloys with a low Cu concentration and showed that Z is 1.14. Applying this model, the average magnetic moment in $Ni_{50}Cu_{50}$ is calculated to be $0.030\mu_B$. Our result is somewhat larger than the calculated value. It is well known that $M_s(0)$ in Ni–Cu alloys deviates from this linear relation when the Cu concentration is higher than 40 at.%. As mentioned above, this has been a source of puzzlement and controversy for a long time.

The present experiment shows that $M_s(0)$ and T_c for the undeformed $Ni_{50}Cu_{50}$ sample are in good agreement with the results of Ahern *et al* (1958), despite the fact that the experimental methods are different. Furthermore, the intrinsic magnetic changes in $M_s(0)$ and T_c occur during plastic deformation. Generally, plastic deformation has three effects on the magnetic properties of alloys, namely the effect of the strain field of the dislocations, the effect of phase transformation due to the strain field and the effect of antiphase boundaries (APB) and stacking faults. In the following, we shall discuss these three effects.

(1) Dislocations nucleate by applying an external stress, which has an influence on the structure-sensitive properties such as initial susceptibility, reversible susceptibility and coercive field. These influences are observed mainly in ferromagnetic metals such as Fe, Co and Ni (Seeger *et al* 1964, Kronmüller 1967). However, the intrinsic change in $M_s(0)$ and T_c cannot be attributed to the strain field around dislocations.

(2) The plastic deformation is sometimes accompanied by a crystal phase transformation. For example, the crystalline structure changes from the face-centred cubic to the body-centred cubic structure when stainless steel of 18–8 type is deformed, and a transition from the paramagnetic state to the ferromagnetic state occurs. Since x-ray diffraction shows that no phase transformation occurs after plastic deformation in Ni–Cu alloy, the phase transformation is not considered to be the reason for the reduction in $M_s(0)$ and the decrease in T_c.

(3) APBs are generated over the glide planes when intermetallic compounds and ordered alloys are plastically deformed; the APB ribbons are produced between superpartial dislocations. The atomic arrangement in an APB is different from that of the ordered state, which certainly alters the magnetic interaction. Accordingly, the magnetic state along the APB ribbons is different from that apart from the APB ribbons.

The magnetic transition due to plastic deformation in general can be explained from the viewpoint of the atomic configuration in the vicinity of the APB between superpartial dislocations (Takahashi and Ikeda 1983). This explanation has been experimentally confirmed in ordered alloys and intermetallic compounds (Takahashi *et al* 1990, 1991, 1994). The decreases in $M_s(0)$ and T_c on plastic deformation of the $Ni_{50}Cu_{50}$ alloy should be explained by the same model as the other ordered alloys and the compounds. In other words there exists an atomically ordered structure in the Ni–Cu alloy, since an APB is produced only in atomically ordered alloys.

The existence of an atomically ordered phase seems a plausible explanation for the deviation of $M_s(0)$ from the Slater–Pauling curve in Ni–Cu alloys. The Slater–Pauling curve is obtained on the assumption that the alloys are atomically disordered. The present specimens would be in the partially ordered state and the plastic deformation decreases the value of $M_s(0)$. This result suggests that good agreement of $M_s(0)$ with the Slater–Pauling curve can be expected for a completely disordered Ni–Cu sample. Different experimental results can be expected in the fully ordered state and in the completely disordered state.

A detailed study of the relationship between the magnetic properties and the dislocation density including the existence of the atomically ordered structure is in progress.

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