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Spin-glass-like behavior in Cu/Ni multilayered films

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A detailed magnetization study of Cu/Ni multilayered films made by sputtering is presented, and it is shown that these films are "reentrant" systems and not ordinary ferromagnets as often assumed. The transition from the spin-glass phase to the ferromagnetic phase is identified by the temperature T_p at which a peak in the dc magnetization occurs. This peak is seen in all the samples and T_p decreases as λ (bilayer thickness) decreases. The ferromagnetic region between T_p and the Curie temperature T_C is also identified.

INTRODUCTION

Cu/Ni is a prototype multilayered system for magnetic studies since Cu and Ni have the same crystal structure and they form solid solutions across the entire composition range. This system has been studied extensively[1–9] in the last few years and was often described as an ordinary ferromagnet. Zheng *et al.*[2] measured the saturated magnetization M_s of Cu/Ni using a magnetic field of 10 kG and found that M_s decreased as the periodicity λ (thickness of Ni+thickness of Cu) decreased. They also found that M_s decreases with increasing temperatures just as in a ferromagnet and the Curie temperature T_C decreases as λ is lowered. Ac magnetization measurements were conducted on Cu/Ni by Zhou *et al.*[9] They concluded that their system is a ferromagnet and they suggested that the Ruderman-Kittel-Yosida (RKKY) interaction exists between Ni layers through the nonmagnetic Cu layers. Swiatek *et al.*[10] studied the exchange interaction between two similar ferromagnetic films of $Ni_{80}Fe_{20}$ across a Cu film layer by light scattering from spin waves. The thickness of Cu was varied between 0 and 50 Å. They also concluded that the RKKY interaction exists through the Cu film for film thickness up to 12 Å.

In recent studies of Cu/Ni (Refs. 11 and 12) with $\lambda \approx 60$ Å, time dependence of the magnetization has been reported. Recently,[13] we have made dc magnetization measurements at low fields for Cu/Ni multilayer films. We observed a peculiar behavior, namely, a peak in the magnetization as a function of temperature and irreversibilities. The temperature T_p at which the peak in the magnetization occurred decreased as λ was varied from 60 to 6 Å. This is reminiscent of the spin-glass behavior in thin Cu-Mn alloy system where Kenning, Slaughter, and Cowen[14] found that the temperature of the peak in the dc magnetization (called T_g) decreased with decreasing the film thickness. T_g was also determined from the peak location in the dc magnetization measurements.

In this work, we have extended our study on Cu/Ni multilayered films to a much thicker layer ($\lambda = 100$ Å) to see whether the same magnetization behavior that we found for smaller λ persists. In addition, more detailed magnetization measurements have been conducted for all samples with different bilayer thickness λ.

RESULTS AND DISCUSSION

The Cu/Ni multilayer samples used in this study were prepared by a UHV sputtering machine. As described earlier,[13] the system was pumped down to a base pressure

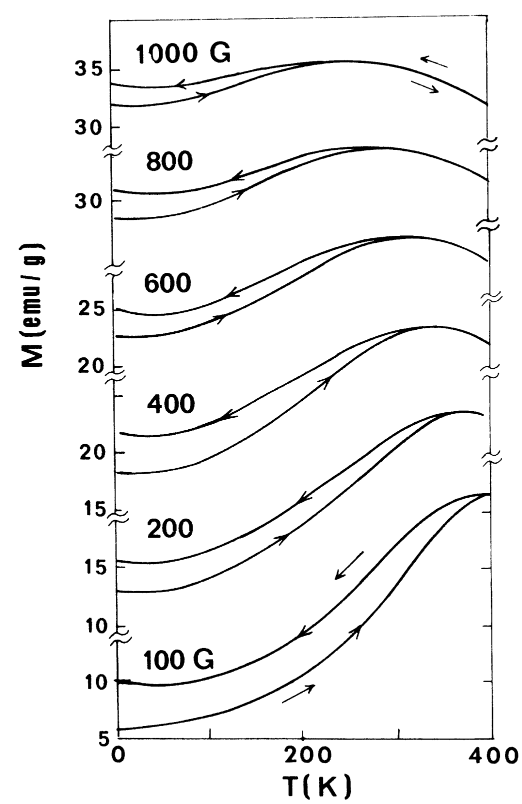

FIG. 1. Magnetization M vs temperature T for $\lambda = 100$ Å in various applied magnetic fields. The arrow to the right represents the zero-field-cooled curve while the arrow to the left represents the field-cooled curve.

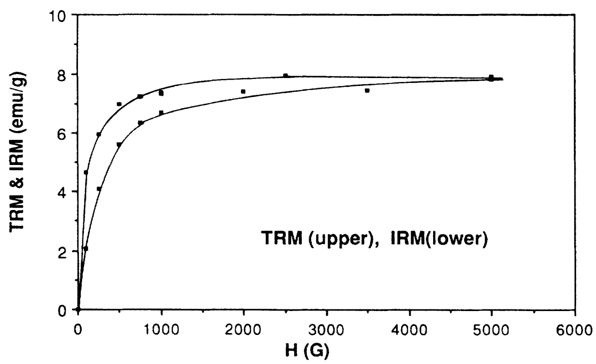

FIG. 2. The thermoremanent magnetization (TRM) vs the cooling field; and the isothermal remanent magnetization (IRM) as a function of the applied field.

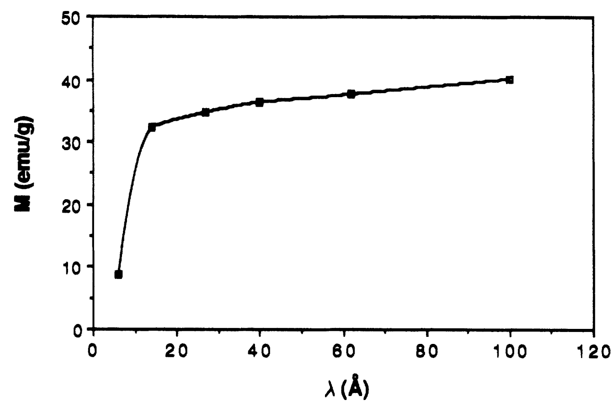

FIG. 4. The magnetization at 10 kG applied field vs λ.

of $\sim 3\times 10^{-9}$ torr before admitting the ultrahigh-purity Ar gas to the chamber. The Ar gas was further purified by passing it first through a cold trap at 100 K to freeze out water vapor and then through a gas purifier to remove impurities such as O_2 and N_2 by reaction with a hot Ti-based alloy. A computer-controlled substrate holder plate moved the substrate quickly between the Cu and the Ni sources. In each sample, the Cu layers have the same thickness as the Ni layers and the total thickness of the sample was 5000 Å.

For the $\frac{50}{50}$ ($\lambda = 100$ Å) sample we show in Fig. 1, the changes of the magnetization M in various applied fixed fields H as the temperature T is raised from 50 to 400 K (represented by the arrow to the right). The sample was initially cooled in zero field. As shown, every curve exhibits a peak that broadens and shifts to a lower T with increasing H. When the sample is subsequently cooled in the same field H, M rises at first along the same curve until it separates itself approximately at the peak position, then it comes down as we continue the cooling all the way to the lowest temperature. (This cooling process is indicated by the arrow to the left.) The behavior of the magnetization curves in Fig. 1 resembles that of spin

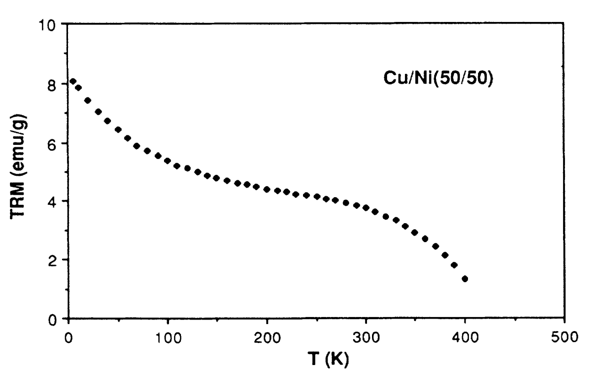

FIG. 3. The saturated thermoremanent magnetization as a function of temperature for $\lambda = 100$ Å sample.

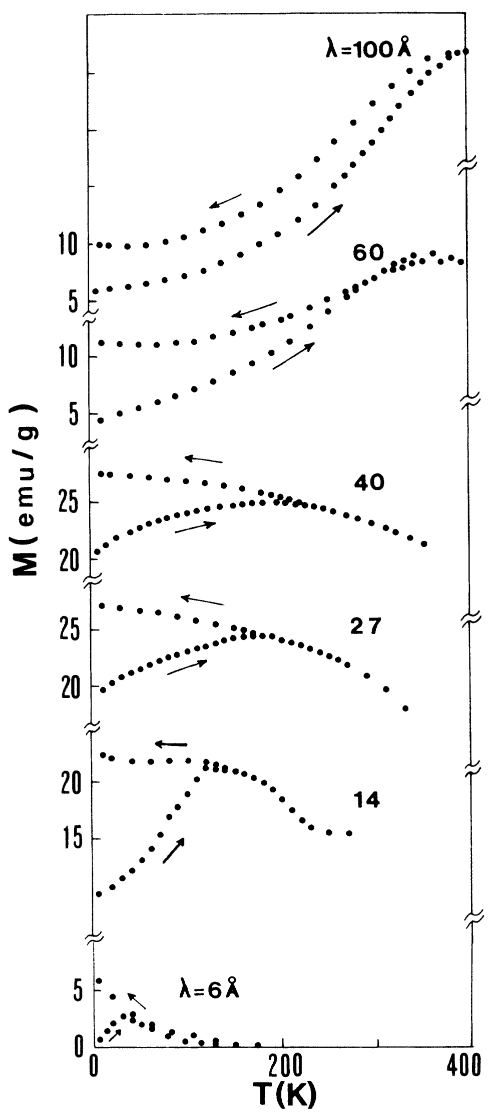

FIG. 5. Magnetization vs temperature of Cu/Ni multilayered films of different layer thickness in 100 G applied field. Both the zero-field cooled curves and the field-cooled curves are shown.

glasses with one exception viz., the temperature at which the peak occurred is relatively high. For the 100 G applied field, it seems that the peak temperature is >400 K and we know of no spin-glass system with such a high transition temperature.

In different experiments, the sample ($\lambda = 100$ Å) was cooled in various cooling fields *H*(cool) to 5 K. The field at 5 K was then lowered from each *H*(cool) to zero, and the thermoremanent magnetization (TRM) was measured. The TRM versus *H*(cool) is plotted in Fig. 2. In the same figure we also show the isothermal remanent magnetization (IRM) versus the applied field *H*. The IRM was obtained by cooling to 5 K in zero field before applying a field *H*, then *H* was turned off and the remanent magnetization (IRM) was obtained. The IRM rises and bends over until it approaches the saturation value of the TRM from below. Similar TRM and IRM behavior was observed for bulk Ni-Mn spin-glass system.[15]

The variation of the saturated value of the TRM with temperature was measured from 5 to 400 K for the $\lambda = 100$ Å sample, and it is shown in Fig. 3. The TRM basically decreased with increasing temperatures and it appears to vanish at ~425 K. This is approximately the temperature at the peak position of *M* versus *T* curve at 100 G applied field, the lowest field used in this study (Fig. 1). Indeed it is obvious from that curve that the peak position would be >400 K (we only could go to 400 K). The fact that the TRM vanishes at a temperature that is approximately equal to the temperature T_p at which the peak in the magnetization occurred, indicates that the peak in the magnetization corresponds to a phase transition. The peak in the magnetization, the reduction of *M* upon cooling, irreversibilities, and the shape of TRM and IRM mimic a spin-glass behavior. However, the magnetization value of the peak at 100 G applied field is relatively high (~27 emu/g) and it is 40 emu/g (see Fig. 4) at 10 kG field. This represents a rather large percentage of the saturated magnetization of pure Ni (55 emu/g). This leads us to believe that this sample probably has a mixed state of ferromagnetism and spin glass below T_p.

The magnetization *M* versus *T* at 100 G applied field of all samples studied of different λ is shown in Fig. 5. In all

TABLE I. Shows the temperature T_p at which the peak in the magnetization occurred of the Cu/Ni samples of different λ, and, the corresponding Curie temperature T_c taken from Ref. 6.

λ (Å)	T_p (K)	T_C (K)
6	30	170
14	120	400
27	182	500
40	200	520
60	357	545
100	425	580 (extrapolated)

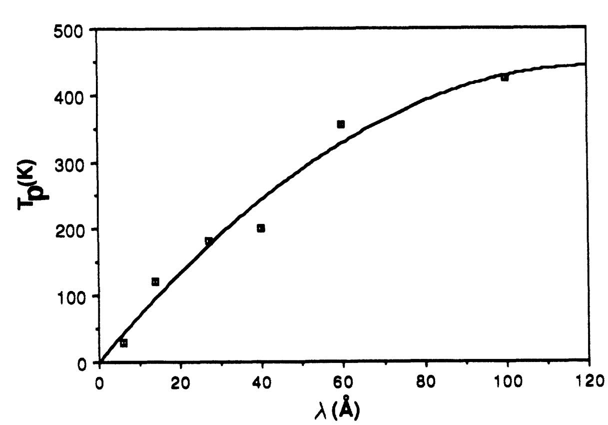

FIG. 6. The temperature that corresponds to the peak in the magnetization at 100 G applied field is plotted against λ.

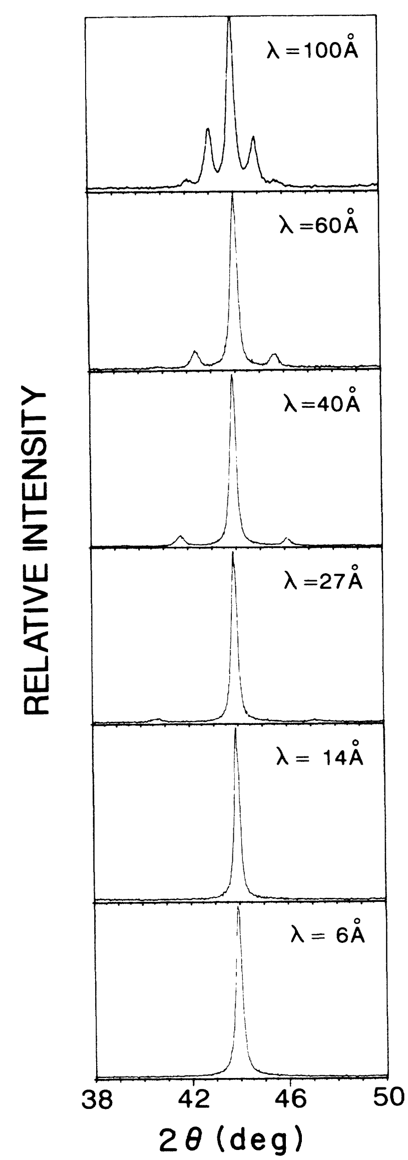

FIG. 7. X-ray diffraction of all our Cu/Ni multilayered samples of different layer thickness.

cases the sample was cooled in zero field to 5 K, then a field of 100 G was applied and the magnetization was measured during heating and then cooling. All the samples exhibit a peak in the magnetization and thermal hysteresis. The temperature T_p at which the magnetization peaked is plotted in Fig. 6 as a function of λ. It is seen that T_p decreases as λ is decreased from 100 to 6 Å. In addition to the systematic behavior of T_p with λ, the field cooled (FC) part of the curves in Fig. 5 also changes systematically with λ. For the thinnest λ, the FC curve rises rapidly with decreasing the temperature from T_p to 5 K, while it rises slowly for $\lambda=14$, 27, and 40 Å and it comes down for the thicker λ, namely for $\lambda=60$ Å and $\lambda=100$ Å. Zheng *et al.*[6] measured the Curie temperature T_C of Cu/Ni multilayered films as a function of λ. These temperatures are listed in Table I along with T_p values (the temperature at which a peak in the magnetization occurred). As shown T_p is considerably smaller than T_C for all thicknesses. This indicates that T_p does not represent a transition from ferromagnetism to paramagnetism. We think that T_p represents a transition from a ferromagnetic phase to a spin-glass-like phase upon decreasing the temperature. Such systems became to be known as "reentrant" systems.

X-ray diffraction studies of our samples are shown in Fig. 7. For $\lambda=6$ and 14 Å, the lack of satellites and the presence of a single sharp (111) crystalline reflection at the average reciprocal lattice spacing for fcc Cu and fcc Ni is indicative of substitutional alloy without chemical modulation. For thicker λ, the satellites appeared and their relative intensities to the central (111) peak increased as λ increased signaling the formation of thicker Cu and Ni layers. It is rather interesting to have a spin-glass-like behavior in all our samples regardless of the thickness of the individual layers. For the $\lambda=6$ Å sample, the peak in the magnetization probably resulted from the alloying of Cu and Ni. Indeed Cu-Ni disordered alloy in the bulk form is known to have a SG-like behavior.[16] So it is not surprising to see a peak in the magnetization for $\lambda=6$ Å sample or even for $\lambda=14$ Å sample. As the Ni layer increases in thickness (as λ increases), it probably competes with the interfacial region (alloyed region). When the Ni layer becomes relatively thick, the interfacial region becomes less important. Despite that fact, as we have shown, the $\lambda=100$ Å sample for example, still have a spin-glass-like behavior. One might speculate that the RKKY interaction between Ni layers through the Cu plays an important role in determining the magnetic properties for samples with large λ.

CONCLUDING REMARKS

Our study has shown that Cu/Ni multilayered films are not ordinary ferromagnets as often assumed up to at least $\lambda=100$ Å. These films appear to have a reentrant phase where the system enters a spin-glass-like phase from a ferromagnetic phase upon cooling. The transition temperature, T_p, from ferromagnetism to the reentrant phase was identified as the temperature at which a peak in the low-field magnetization occurred, while the Curie temperature T_c, given in Table I, represents the transition from ferromagnetism to paramagnetism. The spin-glass-like behavior in this system may be due to two effects, the alloying at the interfaces and the RKKY interaction between the Ni layers through the Cu layers. The former is probably more important for small λ while the RKKY interaction may dominate for thicker λ's.

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