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Magnetic Properties of Multilayer Films of FeNi-Mn-FeNiCo and of FeNi-Mn

O. MASSENET, R. MONTMORY, AND L. NÉEL

Abstract—Experiments made on multilayer evaporated thin films with a structure FeNi-Mn-FeNiCo and FeNi-Mn exhibit strong evidence of a ferromagnetic-antiferromagnetic exchange coupling between the ferromagnetic film of FeNi and a transition layer at the FeNi-Mn interface obtained by mutual diffusion of Mn and FeNi at 300°C. The surface coupling energies measured may be as much as 0.3 erg/cm².

INTRODUCTION

THE PHENOMENON of coupling between the magnetizations of two ferromagnetic films separated by a nonmagnetic metallic layer of suitable thickness, have been studied and the experimental results obtained have been described elsewhere [1]. Since the coupling phenomenon which one observes is sensitive to the nature of the intermediate material, samples had been produced using a variety of intermediate materials. Among them have been films with the structure FeNi-Mn-FeNiCo (19 per cent Fe, 81 per cent Ni — Mn — 13 per cent Fe, 52 per cent Ni, 35 per cent Co).

EXPERIMENTAL CONDITIONS

The films are produced by thermal evaporation in vacuum in the presence of a continuous magnetic field of 100 Oe, which induces an uniaxial anisotropy in the non-magnetostrictive ferromagnetic layers; the easy directions of magnetization of these layers are parallel. The thicknesses of each layer is controlled during evaporation by a quartz oscillator. The pressure during evaporation does not exceed 10⁻⁵ torr, and the evaporation rate is approximately 30 Å/s. The samples are evaporated onto glass substrates, 18 mm in diameter, heated to 280°C.

Magnetic studies are performed primarily with the aid of double Kerr magneto-optic apparatus which permits the hysteresis loops of each of the two magnetic layers to be shown separately on the screen of an oscilloscope; it is operated with an alternating magnetic field of 50 c/s. The easy direction loops are observed at room temperature with the sample in air. A second Kerr apparatus permits the observation of the hysteresis loop of one side of the sample while the sample is in a vacuum chamber where its temperature may be varied continuously between -180°C and 300°C.

EXPERIMENTAL RESULTS

Observations at Room Temperature

For very thin layers of manganese, below 150 Å, we observe a coupling between the magnetizations of the FeNi and FeNiCo films; this coupling is completely analogous to that observed with samples using intermediate layers of chromium, palladium, or silver. This is no longer measurable for thicknesses of 150 Å or greater.

Superimposed upon this coupling is also observed either for the FeNi or for the FeNiCo, a displacement of the hysteresis loop, varying with thickness of the Mn layer, but independent of the magnetic state of the other film and the applied field excursion (less than 50 Oe). This displacement may be interpreted using a fictitious coupling field h acting on the ferromagnetic film; the direction of h is parallel to the direction of the field applied to the sample during evaporation and cooling. Figure 1 shows the values of the displacement of the FeNi loops as a function of the thickness e of the Mn layer for a series of samples prepared on similar conditions, and for which the thicknesses of the FeNi and FeNiCo films are always 1800 Å. Above a certain value of Mn thickness, 250 Å, the displacement no longer depends directly upon the thickness and remains 10 Oe. By analogy with the experiments of Meiklejohn on fine particles of oxidized cobalt [2], one suspects that the observed unidirectional anisotropy of the FeNi arises from an exchange coupling between the magnetic film and a transition layer at the FeNi-Mn interface which is anti-ferromagnetic at room temperature. This transition layer may be produced the mutual diffusion of Mn and FeNi.

One notices that the measured value of h is then related to surface energy of coupling E between the two films experiencing this exchange interaction by the relation

$$E = h J e$$

where J is the spontaneous magnetization (900 emu) of the FeNi layer and e is its thickness (1800 Å). Thus, a displacement of 10 Oe corresponds to a surface coupling energy of $E = 0.16$ erg/cm².

One may attribute a displacement of the loop of the FeNiCo, which is similar but always smaller (< 1 Oe) to the same origin. Further discussion will be limited to the FeNi.

We note that J. S. Kouvel [3] has demonstrated the existence of composition of the type (Fe_x Ni_{1-x})₂ Mn which are antiferromagnetic in the disordered state with transition temperature varying with composition between -70°C and +250°C.

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Fig. 1. Variations of FeNi loop displacement with thickness e of Mn. Thickness of FeNi layer 1800 Å.

(a)

(b)

Fig. 2. (a) Photograph showing the variations of the FeNi loop displacement with temperature from 100 °C to 200 °C. (b) Curve showing the variations of the FeNi loop displacement with temperature varying from -180 °C to +300 °C.

Fig. 3. Curve showing the temperature variations on the FeNi loop displacement. (a) Before annealing. (b) After one annealing. (c) After two annealings.

In order to verify the hypothesis of the existence of a ferromagnetic-antiferromagnetic coupling we have produced samples with the structure FeNi-Mn-SiO, the SiO film being necessary to protect the Mn layer against chemical attack by the air. With these samples is also observed a displacement of the loop of the FeNi of the same sort observed with the samples FeNi-Mn-FeNiCo. Furthermore, after having found that at room temperature it was impossible to change the sense of the displacement or to magnify its amplitude with fields of more than 3000 Oe, certain samples have been annealed at 300°C for varying lengths of time and cooled slowly (three hours) to room temperature in the presence of a constant magnetic field parallel to the easy directions of the samples.

For example, one sample was annealed at 300°C for 15 hours and then cooled in the presence of a 10-Oe continuous field whose direction was opposite to that in which the evaporation was carried out. The hysteresis loop of the FeNi layer photographed before and after annealing shows an increase in the displacement value from 6 to 25 Oe and a reversal in the sense. The displacement direction is thus directly related to the direction of the magnetic field in which the sample is cooled from 300° to room temperature, or (which comes to the same thing in this experiment) to the direction of the spontaneous magnetization of the FeNi during cooling.

The coupling increase on annealing is thought to be related to an increased diffusion of Manganese into the ferromagnetic compound while the reversal of displacement direction during the annealing is a result of the Néel temperature of the antiferromagnetic layer being less than 300°C. When, on cooling, the sample reaches this temperature, the spins of the antiferromagnetic compound begin to arrange themselves in such a way that, at the interface of the ferromagnetic and antiferromagnetic zones, there is minimum exchange energy between the spins of the two zones. When lower temperatures were reached, the antiferromagnetic spins were blocked and they thus induced the FeNi unidirectional anisotropy shown by the loop displacement.

Variation of the Displacement with Temperature

Using a Kerr effect apparatus under vacuum, a systematic study of the variation of FeNi loop displacement for temperatures ranging from -180° to 300°C was carried out. Figure 2(a) and (b) shows the typical variation with temperature of the FeNi loop displacement for a particular sample.

A maximum displacement is observed at about 90 to

100°C followed by a rapid decrease for temperatures greater than 100°C. At about 180°C, the decrease is less rapid and all coupling disappears between 250 and 300°C, depending on the particular sample. For temperatures less than 100°C, a slow reversible decrease in the displacement is observed on lowering the temperature. Furthermore the displacement is independent of the amplitude of the ac field used to observe the FeNi hysteresis loop, at least for amplitudes less than 50 Oe. For temperatures greater than 100°C, the displacement is independent of the applied field amplitude; however, the behavior of the samples (on heating and on cooling) is not the same. The curves given were obtained on heating.

If the sample is left to cool from 300°C with its easy axis parallel to an ac field strong enough to affect the FeNi magnetization, no loop displacement appears. On the contrary, it has been shown that the loop displacement does appear upon cooling in the absence of all external magnetic fields, if the FeNi film is saturated. This results because the exchange interaction between the ferromagnetic and antiferromagnetic layers determines the antiferromagnetic spin orientation, and not their interaction with external fields. After the sample is cooled to room temperature the displacement is greater than it was before heating; its direction can be interpreted using a fictitious coupling field parallel to the spontaneous magnetization of the FeNi during cooling. Figure 3 shows the variation of the coupling strength for a sample subjected to three successive cycles of cooling and heating to 300°C. Each time, the sample temperature was between 250° and 300°C for about an hour.

CONCLUSION

Although it is difficult at the moment to explain the existence of a maximum displacement at about 90°C, the general indication of the experimental results obtained points to the existence of a ferromagnetic-antiferromagnetic-type coupling between the FeNi layer and the transition layer obtained at the FeNi-manganese interface by the diffusion of manganese in the FeNi at temperatures of about 300°C. The surface coupling energies obtained can be as much as 0.8 ergs/cm².

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