An Investigation of Giant Magnetoresistance(GMR) in Granular Thin Films

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Summary

Digital storage technology, driven by the ever increasing need for higher density and faster processing, has reached levels of several gigabits/square inch capacity. One of the basic limitations in achieving denser hard drives in computers is the read head which reads the stored information. Even smaller heads with higher sensitivity are becoming absolutely essential. In recent years the phenomenon of giant magnetoresistance(GMR) in magnetic multilayers, granular films etc., have shown great potential in reaching the above goal. However, these new thin film devices have not reached their optimum potential. This project is aimed at investigating this new effect in granular thin films prepared by an as yet unexplored method, namely, co-evaporation. The results obtained are promising for Ag_2Co films. Certainly more research is needed to reach higher goals.

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Giant Magnetoresistance in Granular Thin Films

As digital storage technology is reaching densities of gigabits per square inch level, it is becoming increasingly important to find ways to have read heads capable of retrieving information. Several new phenomena are being explored in the past 4-5 years.

The phenomenon of giant magnetoresistance(GMR) was investigated in granular thin films. The aim of this project was to understand, study and optimize GMR in co-evaporated granular thin films which has not been reported earlier. The change in resistance of the sample in an applied magnetic field was measured to obtain the magnetoresistance(GMR). In order to achieve the goal, various film compositions like Ag_2Co , $Ag_{60}Co_{40}$, Ag_3Fe, and $Ag_2Ni_{0.2}Fe_{0.8}$ were prepared, and the effect of heat treatment on these films were studied. The best results were obtained in Ag_2Co films, a GMR of 6.22% at room temperature. In the following report, results from this research effort is presented and discussed.

Introduction

Recently, due to technological importance, many scientists and engineers are motivated to study the giant magnetoresistance effect(GMR) in ferromagnetic multilayer thin films. Thin films with a GMR effect could be used as magnetic read heads in the recording industry[1,2].

In the past, and currently, conventional inductive heads were used in magnetic recording industry. These heads are proving to be impractical as the storage density has increased tremendously. Anisotropic magnetoresistance(AMR) thin film heads are recently being used to obtain higher areal density. AMR heads rely upon the change in resistance of a sense layer($\Delta R/R$) of a ferromagnet(FM) in response to magnetic field parallel, and transverse to the current in the sensor. The AMR heads were first brought into the market by IBM in 1991, and now all IBM drives utilize these heads[2]. The main problem with AMR heads is that they have limited sensitivity. Relative to AMR heads, GMR heads have a 25-50% improvement in sensitivity[2]. In the future, GMR heads will find their way to record larger amounts of information on a hard drive, reaching > 10 Gbits/square inch.

A magnetoresistance effect is the relative change in resistivity ($^{\Delta R}/_{R}$) of a material in an external magnetic field. This magnetoresistance effect is usually low and less than 1% in most materials. But there are some specially tailored materials, like magnetic multilayers(FM/normal metal layers repeated)[3,4] and granular magnetic systems(non immiscible mixture of FM and normal metal)[5-9], that show a giant magnetoresistance effect(GMR) which is found to be several percent to orders of magnitude change. This GMR effect depends on both electron mean free path(l), interface conditions in multilayers, and the magnetic particle size especially in granular systems. The value of l is determined by spin dependent electron scattering on ferromagnetic particles. Granular thin films are prepared in different ways at ambient, lower, or higher temperatures and then are annealed. This annealing process separates the magnetic substances from the non-magnetic substances which permits the magnetic particles to grow larger in the plane[2]. In the past, only multilayer thin films were used in studying the GMR effect, until recently, when it was discovered that granular thin films could also give rise to significant GMR effects. Granular systems are simpler to fabricate[10], although the GMR effects are lower compared to multilayers. In the granular thin film studies reported so far, we did not find any report on co-evaporative techniques to create thin films. Co-sputtering is the common deposition technique for growing granular thin films followed by other workers[3-9,11]. We decided to explore co-evaporation technique to grow Ag-Co, Ag-NiFe and Ag-Fe granular thin films, and our goal was to achieve a high value of GMR in these films as deposited or by annealing at various temperatures.

Experimentation

Thin films of Ag_2Co, $Ag_{60}Co_{40}$, $Ag_2Ni_{0.8}Fe_{0.2}$ and Ag_3Fe were prepared using co-evaporation techniques. A glass substrate was used to grow the granular films. The silver(Ag) was placed in a tantalum(Ta) boat and cobalt(Co) was placed in an electron gun(e^- gun) crucible inside the vacuum. The initial background pressure in the chamber was =1.3 x 10^{-4} Pa. A high current was passed through the Ta boat containing Ag which melts the Ag material(melting point=1233.5K). Thus,the co-evaporation of Ag and Co took place. The amount of heat produced was directly dependent on the amount of current,time and resistance of the Ta boat($Q=I^2Rt$). Enough heat was provided for the Ag to melt and evaporate. Since Co has an even higher melting point(1718K), it is more difficult to melt Co and we resorted to using an e^- gun. The e^- gun contains a tungsten filament which supplies a stream of electrons accelerated towards the Co source by a high voltage(3,000V). Because the electrons have great energy due to this high voltage acceleration, when incident on Co, the metal melts and evaporates. The amount of Ag or Co deposited can be precisely controlled by film thickness controllers called quartz crystal monitors(QCM). (QCM works on the principle of changing the resonant frequency of a single crystal quartz plate,which is part of an oscillator circuit, by the deposited film mass.) By this technique one can produce films of any chemical composition and thickness. The glass substrate could be maintained at any temperature from 77K to 850K. Films of Ag_2Co were prepared holding the substrate either at 77K or 395K(+/- 2K). This was done to see the effects of substrate temperature on GMR. Films of silver permalloy($Ag_2Ni_{0.8}Fe_{0.2}$), $Ag_{60}Co_{40}$, and Ag_3Fe were also prepared in separate evaporation runs, using the same techniques as mentioned above.

After the films were made, different annealing processes were adopted. To do this,the thin films were placed in a copper box, which was placed next to a heater, in a vacuum chamber. An external heater controller maintained any desirable temperature of the film. A thermocouple wire, placed in the copper box adjacent to the specimen, indicated the temperature(+/- 1K) of the film. The thermocouple puts out a small voltage which can be read with a voltmeter,and is directly proportional to the temperature. Standard voltage to temperature conversion tables are available. The first annealing temperature was chosen to be 367K, and based on these results, other annealing temperatures were chosen to achieve greater GMR results. Table I below shows film preparation and annealing conditions for various samples.

Table I

Specimen	Substrate Temp(K)	Annealing Temp(K)	Time of Annealing
Ag_2Co	77	367	8min,25s
Ag_2Co	77	502	10min,15s
Ag_2Co	395	593	13min
Ag_2Co	77	593	13min
Ag_2Co	395	543	20min,15s
Ag_2Co	77	543	20min,15s
Ag_2Co	395	518	19min,15s
Ag_2Co	77	518	19min,15s
Ag_2Co	395	453	13min,40s
Ag_2Co	77	453	13min,40s
Ag_2Co	77	408	4min
$Ag_{60}Co_{40}$	77	573	5min,30s
$Ag_{60}Co_{40}$	77	673	5min
$Ag_{60}Co_{40}$	77	453	5min,10s
$Ag_2Ni_{0.8}Fe_{0.2}$	77	518	19min,15s
$Ag_2Ni_{0.8}Fe_{0.2}$	77	453	13min,40s
Ag_3Fe	77	453	5min,30s
Ag_3Fe	77	573	5min,30s
Ag_3Fe	77	673	5min

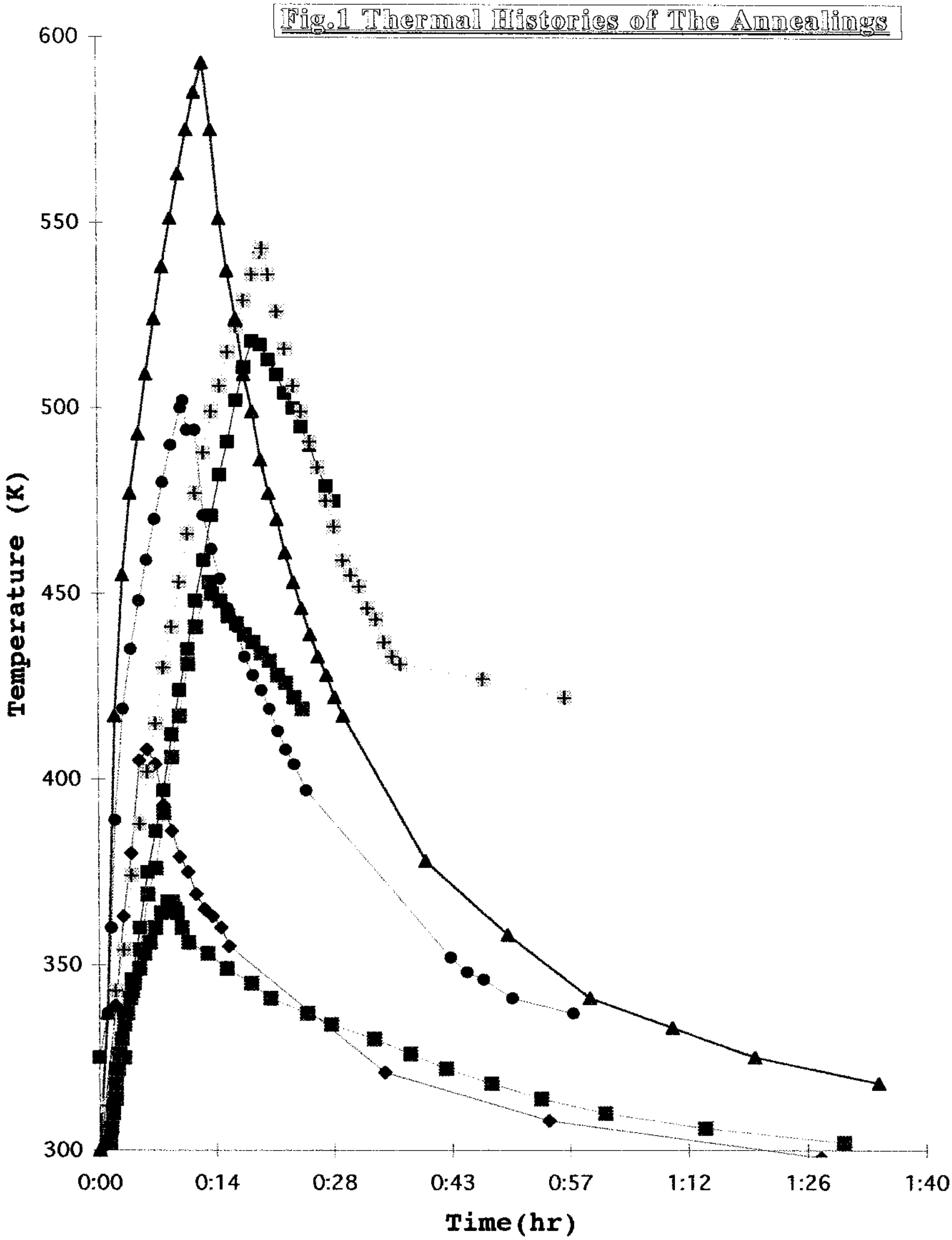

Fig.1 Thermal Histories of The Annealings
Temperature (K)
600
550
500
450
400
350
300
0:00
0:14
0:28
0:43
0:57
1:12
1:26
1:40
Time(hr)

The thermal history graphs show that the films took a long time to cool down. The reason for this is that the films were enclosed in a vacuum chamber, and there was no air present. The only way the films could physically cool down was by radiation, and this was the reason for the long cooling period.

Once films had been annealed and cooled down to room temperature(298K), resistivity measurements as a function of magnetic field were carried out. The annealed thin film, with four attached copper wires(displaced from each other), was placed on a probe and then the probe was in turn placed in an external magnetic field. An ac bridge was used to measure the resistance of the film by the "four terminal technique". In this method, a small ac current is passed through two outer leads and the ac voltage is detected using two inner leads(which are placed very near the current leads). This allows a clean resistance measurement of the sample without the contribution from the test leads. The resistivity was first recorded at zero field(R_I). After increasing the field to a maximum of 0.44 tesla, the resistance was then recorded(R_F). The percentage of GMR was then calculated as follows: $(\Delta R/R)*100$, where $\Delta R=R_F-R_I$ and $R=R_I$. These measurements were carried out for all annealed films, at room temperature, and for some films at 77K. Percent changes were recorded.

Results and Discussion

The main goal of the project was to optimize GMR values. The magnetoresistance of the granular film was observed to be dependent on the annealing history of the sample. Subjecting the film to heat treatment had mixed results. Certain intermediate temperatures gave the highest value of GMR. For example, for Ag_2Co film, annealing at about 453 K had the best value compared to other temperatures of annealing(T_a) > or <453K. Fig. 2 below and fig. 3 on following page show GMR values after annealing at various temperatures.

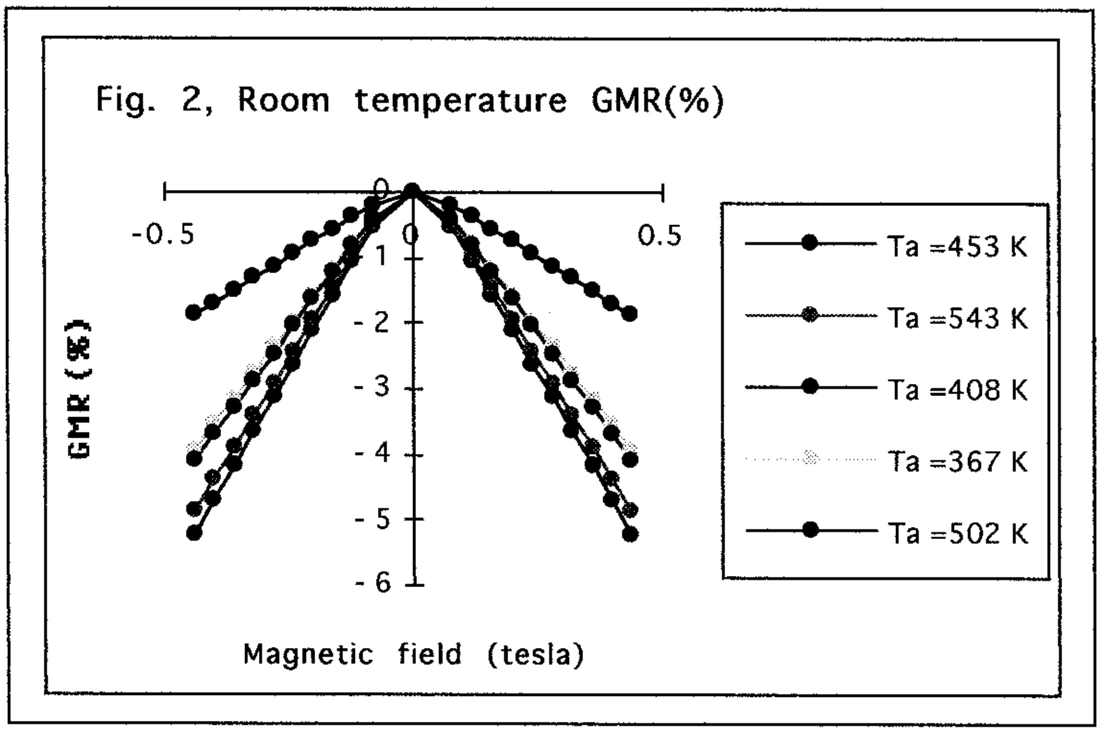

Fig.2. GMR(%) in Ag_2Co films subjected to various heat treatments in a vacuum.

Fig. 3

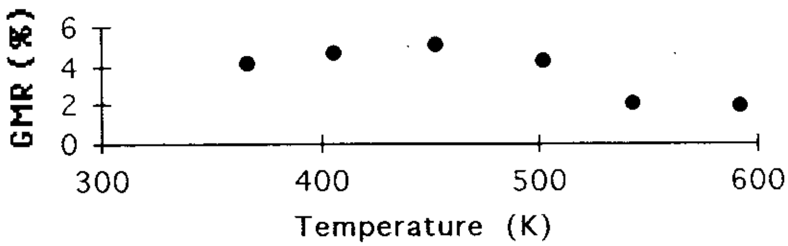

Fig.3 The change in GMR(%) as a function of annealing temperature for Ag_2Co film.

The lowest annealing temperature for Ag_2Co was 367 K and the GMR value obtained here was 4.1%. In order to achieve a higher GMR, higher temperatures of annealing were investigated. Upon increasing T_a to 502 K, the GMR increased to 4.3%. Further increase in T_a resulted in poor values of GMR, in fact reaching only 2%. The optimum annealing temperature was thus observed for these samples to be between 390 K to 500 K. A high GMR of 5.0% was achieved at the above optimum annealing temperature of 453 K.

Another approach to increase the GMR values was to slightly change the composition of Ag-Co films[5-8]. The composition was changed to $Ag_{60}Co_{40}$ and the films were made thicker (100 nm). By annealing to 453 K, a 0.5 % GMR was seen. Just in case this composition had different annealing parameters, for an optimum result, higher temperatures were tried. The film was annealed to 573, and 673 K. Results here were not as good as the previous composition. Only 0.6% and 0.4% GMR was obtained.

Similarly, $Ag_2Ni_{0.8}Fe_{0.2}$ films were also tested for GMR. When T_a=453 K, the GMR was 0.2%. At T_a=493 K, no GMR was seen. This sample did not show potential, in our limited studies, for producing high GMR results. No more experiments were performed.

The third film chosen, Ag_3Fe, was annealed at three different temperatures. The first T_a chosen was 453 K. The magnetoresistance here was low,only 0.5%. Upon increasing T_a to 573 K, a slight improvement was seen with a 0.7% result. In a further attempt to acquire a greater magnetoresistance, the T_a was increased to 673 K. Results here were better, at 1.5%, but the magnetoresistance was still too low.

Ag_2Co was also tested at 77 K to investigate the temperature dependence of the specimen. Fig.4 below shows results from this experiment.

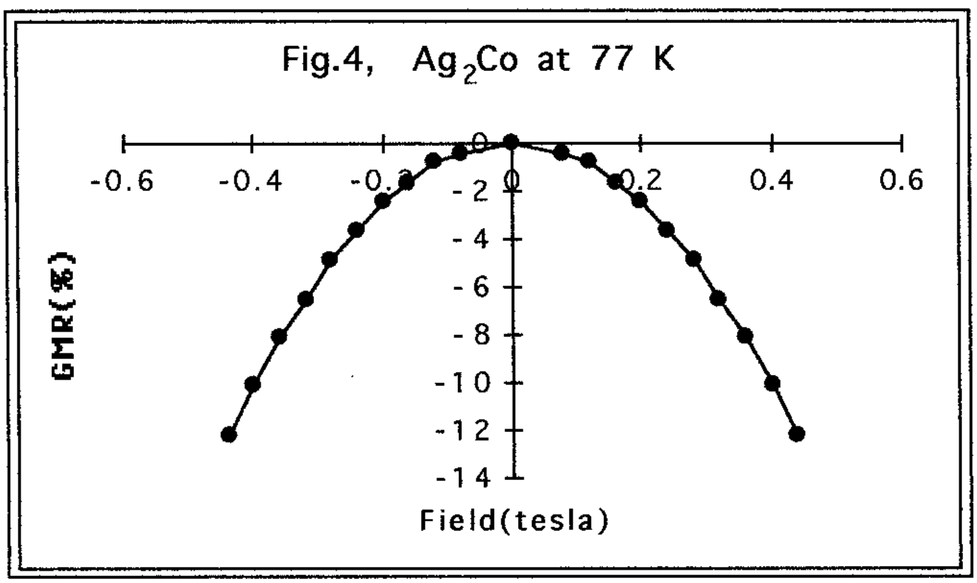

Fig.4 Ag_2Co resistance as a function of magnetic field,at 77K.

Some samples of Ag_2Co were placed in a vacuum dessicator for a period of one month. When one of these samples was tested at room temperature, as prepared, 6.2% GMR was obtained. This was our highest result. Annealing this sample to 453 K did not change the GMR, and it remained at 6.2 %. When the T_a was increased to 573 K, a lower GMR was seen, 4.1%. This sample was also tested at 77 K, and the GMR result was 13.2%. Fig.5 below shows the GMR curve for this sample as prepared.

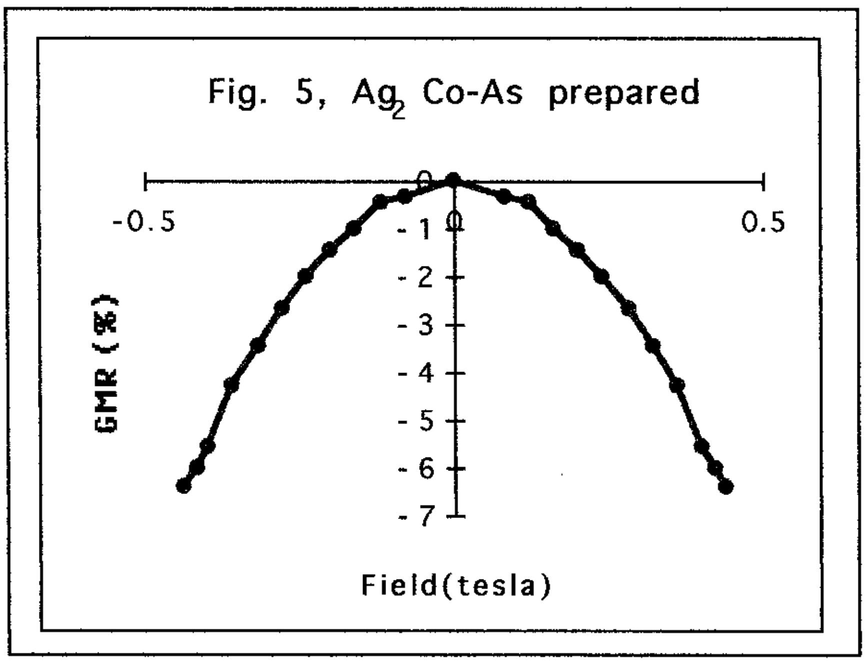

Fig.5 Ag_2Co film(as prepared) resistance as a function of magnetic field for the sample which was stored in vacuum for one month.

Physical processes that might occur during annealing are perhaps as follows. Annealing causes Ag and Co to separate[2].

The Ag, being a rapidly diffusing noble metal, will separate the ferromagnetic Co grains. The experiments with annealing show that it is important to have consistent annealing to produce consistent grain size distributions. Too much annealing will make the grains grow oversized and reduce the GMR effect. With too little annealing, the grains may not grow large enough, therefore lowering the GMR for the specimen.

A simple explanation of GMR in granular films can be provided by the phenomenon of electron scattering[2](which is the source of resistivity in metals). Fig.6 below shows how the scattering of conduction electrons change in different applied magnetic fields. At field(H)=0, electron scattering is high, producing a higher resistance due to the random orientation of the magnetic particles(close to being antiparallel). At higher fields the scattering of electrons is minimal, producing a lower resistance, since all the particles are aligned parallel along the field.

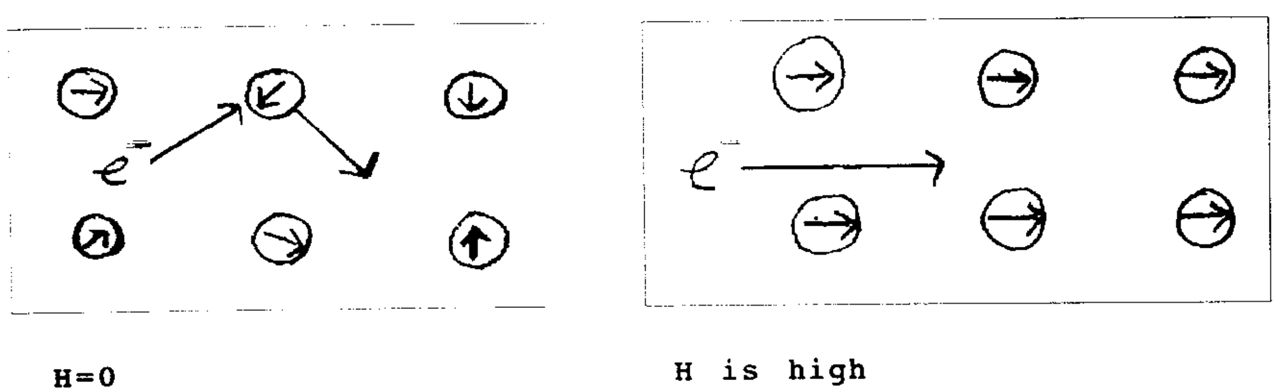

Fig.6 Scattering of conduction electrons at H=0, and when H is high.

Perhaps, the temperature dependence of GMR obtained in our sample can be explained as follows. The ferromagnetic ordering temperature (Curie temperature) of magnetic materials depends on the size of the particles, for small particles[2]. At a lower temperature, the smaller particles may become ferromagnetic, thus producing a larger GMR effect. Using the films at a low temperature may be impractical for technological uses at the present time.

Over 6.2% GMR effect is seen in Ag_2Co sample left in a vacuum dessicator for the period of one month. We have speculations why this might have happened. Ag and Co are insoluble materials. Over this long period of time, the particles may have separated. Possibly, very small clusters of Ag and Co were formed. This separation may have increased the GMR effect.

Conclusion

Technologically important GMR effects in granular thin films were investigated in this research project. Various granular systems(Ag_2Co,$Ag_{60}Co_{40}$,Ag_3Fe,and $Ag_2Ni_{0.8}Fe_{0.2}$) prepared by co-evaporation, were explored to reach potentially useful film composition. It is found that annealing produces significant GMR characteristics in some granular thin films. In Ag_2Co, the highest result(GMR=6.2%) was achieved by leaving a sample in a vacuum dessicator for one month. A high GMR value(5.0%) was also obtained when T_a=453 K. $Ag_2Ni_{0.2}Fe_{0.8}$ films did not show significant GMR before or after annealing treatments. Although freshly prepared Ag_3Fe films did not show much GMR, they did show an increase in GMR as T_a increased. GMR is closely related with spin dependent electron scattering in granular systems with ferromagnetic particles.

Acknowledgements

I would like to thank all those who have helped and supported me throughout my research endeavor. First and foremost, I want to express my deepest gratitude to my mentors, Dr.Jagadeesh Moodera and Dr.Janusz Nowak. Their time, patience, interest, and guidance inspired me to follow through with my research and this Westinghouse project. They taught me all I know about ferromagnetic thin film technology and besides being excellent teachers, I also consider them my close friends. I want to also thank the faculty at Tappan Zee H.S. that helped me keep up with my studies while I was at M.I.T doing research. I'd especially like to thank Mr. Michael Francesco, my science teacher, for his kind help and guidance. Last but not least, I would like to thank my family for their continued faith in me. Without them, none of this would have been possible.

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