行政院國家科學委員會補助專題研究計畫成果報告
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計畫類別：□個別型計畫  □整合型計畫
計畫編號：095-2811-M-006-002
執行期間：94年8月1日至95年10月31日

計畫主持人：梁榮俊
共同主持人：無

本成果報告包括以下應繳交之附件：
□赴國外出差或研習心得報告一份
□赴大陸地區出差或研習心得報告一份
□出席國際學術會議心得報告及發表之論文各一份
□國際合作研究計畫國外研究報告書一份

執行單位：國立成功大學物理系

中華民國95年12月25日

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Preparation of NSC Project Reports
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主持人：黃榮俊 国立成功大學物理系
共同主持人：無
計畫參與人員：黃榮俊、許華書、許峻瑜、李宗勳、顏君乘、涂朝亮、陳宗彥、邱詩航、劉怡君 国立成功大學物理系

一、中文摘要
我們利用雙離子槍濺鍍系統系統化的研究了鈷鐵硼-銣-鈷鐵硼人工反鐵磁系統之磁性耦合變化。我們觀察到適當的離子助鍍電壓可以明顯地增強人工反鐵磁飽和場(從未助鍍的220Oe增強到60伏特助鍍的1000Oe)以及維持鈷鐵硼系人工反鐵磁層的非晶結構，此法提供一個非常簡便的方法去增強人工反鐵磁耦合系統的反鐵磁飽和場以及有利氧化鎂絕緣層發展其紋理結構的基底，對於磁性穿隧接面與磁性隨機存取記憶體的發展有其幫助。

Keywords: dual ion-beam sputter, amorphous, synthetic antiferromagnet, CoFeB

二、緣由與目的
Extensive studies and vigorous development on magnetoresistance random access memory (MRAM) based on synthetic antiferromagnets (SyAFs) and magnetic tunnel junctions (MTJs) have been demonstrated in recent years.

The SyAFs compose of two ferromagnetic (F) layers separated by a nonmagnetic layer (NL). The F layers are ferromagnetically or antiferromagnetically coupled at different NL thickness according to the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction. The crystalline or polycrystalline 3d ferromagnets, such as Fe, Co, NiFe or CoFe, are the most popular ferromagnetic layers in SyAFs. Amorphous 3d ferromagnets, e.g. polycrystalline 3d ferromagnets doping with metalloids like boron, in recent years has gained a new interest to substitute the polycrystalline 3d ferromagnets as the magnetic electrodes of MTJs which demonstrate very large TMR ratio (70% for Al₂O₃ and 470% for MgO insulating layer) due to the improvement of interfacial roughness and MgO (001) texture. However, the use of amorphous 3d ferromagnets in SyAFs shows much weaker coupling strength compared to the polycrystalline 3d ferromagnets.

Developing a technique to enhance the coupling strength of amorphous SyAFs (good magnetic pinned layers) and retain amorphous...
structure (critical for high TMR ratio) is very useful in the application of magnetic sensors and MRAMs.

In this work, we utilize the ion-beam assisted deposition (IBAD) technique to modulate the magnetic coupling of amorphous SyAFs with CoFeB – Ru – CoFeB structure. Possible mechanisms for the variation of magnetic coupling are discussed based on the crystalline texture, presence of pinhole in space layer, and interfacial roughness. This technique provides a convenient route to modulate the interlayer coupling strength for SyAFs and retain the subsequent MgO layer with good textured structure.

SyAFs of Ta / Co_{60}Fe_{20}B_{20} (~2nm) / Ru (t_Ru nm) / Co_{60}Fe_{20}B_{20} (3nm) / glass substrate were prepared by a dual ion-beam sputter system with a base pressure of 5 × 10^{-7} Torr. The IBAD process was achieved by first starting the main sputtering gun with fixed 500V beam voltage and 20mA beam current to sputter Co_{60}Fe_{20}B_{20} (CoFeB) or Ru targets, then additional Ar gas was introduced into the assisted gun under voltage from 0 to 140V to bombard the samples in the process of CoFeB - Ru - CoFeB deposition. During the growth, a magnetic field about 500Oe was also applied to induce the magnetic easy axis. The magnetization curves were measured by vibration sample magnetometer (VSM) at room temperature and post-annealing process was performed at 240°C for 1h under high vacuum (~5 x 10^{-6} Torr) condition. X-ray reflectivity was measured at the wiggler BL17A beamline of National Synchrotron Radiation Research Center in Hsinchu, Taiwan.

### 三、結果與討論

Figure 1 shows the saturation field of magnetization curves as a function of Ru thickness (t_{Ru}) from 1.2 to 1.8 nm. The antiferromagnetic (AF) coupling can be observed in the range of t_{Ru} from 1.2 to 1.5 nm, as shown in Fig. 1. The AF coupled magnetization curve for the t_{Ru} = 1.2 nm sample is shown in the inset (a) of Fig. 1. With t_{Ru} further increases from 1.5 to 1.8 nm, the interlayer coupling transits to F coupled region and the typical magnetization curve is shown in the inset (b) of Fig. 1. In the AF coupled region, the coupling strength for the t_{Ru} = 1.2 nm is almost five times larger than the t_{Ru} = 1.4 – 1.5 nm of the CoFeB – Ru – CoFeB SyAFs. Thus, we focus on the influence of IBAD on interlayer coupling for the SyAFs with t_{Ru} = 1.2 nm in the following discussions.

![Figure 1](image.png)

**Figure 1** The saturation field of the CoFeB / Ru(t_{Ru}) / CoFeB SyAF as a function of Ru thickness of where t_{Ru} = 1.2 to 1.8 nm. The insets (a) and (b) of Fig. 1 respectively show the magnetization curves of antiferromagnetic (t_{Ru} = 1.2 nm) and ferromagnetic coupling (t_{Ru} = 1.8 nm).

Figure 2 displays the magnetization curves of CoFeB - Ru (1.2 nm) - CoFeB under different IBAD voltage from 0 to 140V. For the 0V IBAD SyAFs, the magnetization curve, with a minor loop in low field, has an antiferromagnetically coupled saturation field (AFCSF) of ~200 Oe. The AFCSF of magnetization curves show a significant enhancement up to 1000 Oe with the IBAD voltage increased to 60V. However, the AFCSF for 100V IBAD SyAFs shows a decrease to 440 Oe, which is still larger than the 0V IBAD SyAFs. For further increasing of the IBAD voltage to 140V, the AF coupling disappears and the sample transits to F coupling. The result demonstrates that the IBAD is a useful technique to enhance the AF coupling and to modulate the coupling configuration.
It is noticed that the mechanism underlying for the enhancement of magnetic coupling for SyAFs and the AF to F coupling transition is less studied or understood. Generally speaking, crystalline texture of ferromagnetic layers, presence of pinholes in spacer layer, and interfacial roughness are important factors influencing the magnetic coupling under IBAD process.\textsuperscript{14-16} The variation of crystalline texture of ferromagnetic layers under IBAD process can be inspected directly by the X-ray diffraction spectra and indirectly by the coercivity of magnetization curves. No characteristic diffraction peaks near $45^\circ$ of our CoFeB\textsuperscript{17} can be observed for the 0 and 60V IBAD SyAFs, as shown in the inset of Fig. 2. In addition, the coercivity of the 60 and 100V IBAD SyAFs are almost the same as the 0V samples, as shown in Fig. 3(a). The results clearly indicate that the IBAD processes retain the amorphous structure of top and bottom CoFeB ferromagnetic layers, thus, the possibility of crystalline texture induced enhancement of AF coupling can be excluded. In addition, the presence of pinholes by annealing process in Ru layer has been correlated by observing the change of AFCSF of SyAFs.\textsuperscript{18} Figure 3(b)-3(d) display the magnetization curves of the 0, 60, and 100V IBAD SyAFs annealed at 240°C for 1h. We find that the AFCSF of the 0, 60, and 100V IBAD SyAFs can be retained or enhanced due to the annealing. The results suggest the presence of pinholes in the Ru layer is not likely the most important factor influencing the variation of AFCSF of the SyAFs. The AF to F variation for the SyAFs under IBAD, instead, is more likely related to the change of interfacial roughness between CoFeB and Ru layers, as revealed by x-ray reflectivity measurements. Figure 4 shows the X-ray reflectivity of 0, 60, 100 and 140V IBAD SyAFs and no visual difference on amplitude of oscillation peaks can be observed. Here, we use the parratt32 program to simulate these X-ray reflectivity data.\textsuperscript{19} The layers included to simulate the data contain the bottom CoFeB, Ru, top CoFeB, Ta and natural oxidized Ta.
The simulation curves (solid curves) are shown in figure 4(a) and the analyzing parameters of interfacial roughness and layer thickness are shown in figure 4(b) and 4(c). The top and bottom interfacial roughness as a function of $V_a$ shows a minimum around 60V, as shown in the inset of figure 4(b). The interfacial roughness of the 60V and 100V IBAD samples remain smaller than the 0V SyAFs. We suggest that the change of interfacial roughness due to IBAD process results in the variation of interlayer coupling strength. That is, smaller interfacial roughness leads to larger interlayer coupling strength for the SyAFs, due likely to the weak Neel coupling effect.\footnote{12} Besides, we also find that for the 140V IBAD sample, the bottom interfacial roughness (~4nm) is larger than the Ru layer thickness (~3nm). The result indicates 140V IBAD could etch the CoFeB and Ru layers, leading to a discontinuous Ru layer, and thus the AF coupling transits to F coupling (Fig. 2). Besides the enhancement in antiferromagnetic interlayer coupling of CoFeB-based SyAFs by IBAD process, the formation of the MgO (001) on CoFeB-based SyAFs is also very important because amorphous CoFeB layer can promote the growth of highly textured MgO (001) layer and produce giant tunnel magnetoresistance.\footnote{9,13} Fig. 4(d) shows the Transmission Electron Microscope (TEM) images of a typical MTJ structure, Ru / CoFeB / MgO / 60V IBAD treated CoFeB-based SyAFs / Ru, grown on thermal oxide Si wafers. Obviously, good textured MgO layer (~ 1.5 nm) forms on amorphous CoFeB-based 60V IBAD SyAFs, as revealed by the zoom-in graph in Fig. 4(e). We conclude that the enhancement of AF interlayer coupling and the retaining of amorphous structure of CoFeB-based SyAFs by IBAD is important for the fabrication of MTJs and development of MRAMs.

四．計畫成果自評

We have included the ion-beam assisted deposition method to enhance the antiferromagnetically coupling field of amorphous CoFeB-based SyAFs and retain the amorphous structure of CoFeB layer. The amorphous CoFeB layer is very important for the development of highly textured MgO layer in MTJs because the coherent tunneling process induced by highly textured MgO leads to a giant magnetoresistance ratio. Besides, the enhanced antiferromagnetically coupling field can be a good pinned layer and is useful for MRAM development.

五．參考文獻