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Effect of ion irradiation on the characteristics of magnetic tunnel junctions

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Abstract. The effect of disorder caused by MeV heavy ion irradiation on the tunnel magnetoresistance (TMR) of magnetic tunnel junctions is investigated. Two types of tunnel junctions, viz. Co/Al$_2$O$_3$/Ni$_{80}$Fe$_{20}$ and Co/Gd-doped Al$_2$O$_3$/Ni$_{80}$Fe$_{20}$ are studied. Upon 70 MeV Si ion irradiation at a fluence of $5 \times 10^{11}$ ions/cm$^2$, the undoped junctions show relatively small but irreversible change, while the Gd-doped junctions show a huge reduction, in TMR. In both cases junctions were completely destroyed by 200 MeV Ag ion irradiation at a fluence of $1 \times 10^{11}$ ions/cm$^2$. The results are attributed to the modification of the barrier and the neighboring interfaces caused by the high energy density deposited by incident ions.

PACS. 61.80.Jh Ion radiation effects – 85.30.Mn Junction breakdown and tunneling devices (including resonance tunneling devices) – 85.70.Kh Magnetic thin film devices: magnetic heads (magnetoresistive, inductive, etc.); domain-motion devices, etc.

1 Introduction

During the last decade, Tunneling Magnetoresistance (TMR) in Magnetic tunnel junctions (MTJ) using amorphous Al-oxide tunnel barrier has been widely studied [1,2]. It has also formed the basis for several devices as the solid state magnetic random access memories (MRAM) and magnetic sensors. In the simplest geometry the MTJs consists of two ferromagnetic electrodes separated by an ultrathin insulating barrier. Until recently such MTJs exhibited TMR ratio up to 70% at room temperature using Al$_2$O$_3$ barriers [3]. The TMR ratio is defined as $(R_{UP} - R_{AP})/R_{AP}$ where $R_{UP}$ and $R_{AP}$ are the tunneling resistances when the magnetization of the two electrodes are aligned antiparallel and parallel respectively. The recent demonstration of a high TMR ratio of $>230\%$ at room temperature using MgO as tunnel barriers [4,5] have beyond doubt, increased the feasibility and potential of using such devices for several technological applications. The dramatic decrease of TMR with increasing applied bias is a major impediment in all applications with MTJs. Such a variation with bias is ascribed to several factors as the voltage dependent density of states at the Fermi level [6,7], the quality of the tunnel barrier (I) as well its interface with the ferromagnetic (FM) layer and to magnon excitations at the metal-barrier interface [8].

The effect of doping with magnetic and non-magnetic impurities at various regions of the barrier on the tunneling characteristics has been studied quite extensively [9,11]. However, the sensitivity of the FM-I interface and the density of states in the FM is just beginning to be understood. In this context, the study of MTJs for application in hostile environment such as outer space is quite important.

Ion irradiation is a well-established method to incorporate controlled defects and disorder in materials with homogeneous spatial distribution over a specific sample area. When energetic ions pass through a solid it loses energy either by inelastic collision with the atomic electrons of the solid or by elastic scattering from the nuclei of the atoms of the solid. Whereas the former (electronic energy loss $S_e$) is believed to cause material modifications in the solid or by elastic scattering from the nuclei of the atoms of the solid. A suppression of magnetic interface anisotropy in Co/Pt system [12] and an increase of the same due to chemical ordering in FePt(Pd) [13] alloys has been observed. Using resist masks in conjunction with light ions as He$^+$ or focused Ga$^+$ ion beam [14,15] on similar systems, modifications in the nanometer scale, of the magnetic properties have also been observed. This has found profound potential applications in patterned perpendicular magnetic media for ultra-high density storage. Swift heavy ion (SHI)
irradiation of solids often brings in irreversible changes in materials such as structural changes, interface modification or alterations of the phase composition through the formation of defects [16–19]. In light of this, effect of ion irradiation on the characteristics of MTJs, in particular, the sensitivity of the FM/I interfaces and the ultrathin barrier layer is quite important to study. This will enable one to investigate the interfacial and barrier disorder effects on the magnetic properties, spin transport and tunneling behavior in a controlled manner as a probable step towards making better tunnel junctions for special applications. In a recent experiment, Conraux et al. [18] have studied the effect of light (C, O) and heavy ions (Ni) on an exchange-free and exchange-biased synthetic antiferromagnetic-pinned MTJs. They show that for all ions the TMR decreases without any impact on the overall resistance values.

2 Experimental details

In this letter, we report on the effect of SHI irradiation (Si and Ag) on the junction properties of two types of MTJs, (without any exchange biasing) viz. Co(8 nm)/Al 2 O 3 (1.5 nm)/Ni 80 Fe 20 (10 nm) (MTJ-1) and Co(8 nm)/Gd(0.2 nm)-doped Al 2 O 3 (1.5 nm)/Ni 80 Fe 20 (10 nm) (MTJ-2) for two ion species and energies. We show that both the TMR and the resistance decrease in both types of junction for the lighter ion, whereas for the heavier ion the junctions are completely destroyed.

Details about sample preparation have been discussed in details elsewhere [1]. As mentioned above, we have used an Al 2 O 3 barrier layer thickness of 1.5 nm in both types of junctions: undoped and doped with 0.2 nm of Gd in the barrier. FM films were grown in an applied field of about 100 Oe to create a well-defined coercive field with unidirectional anisotropy. TMR values were obtained from four-terminal measurements of the junction resistance at room temperature (RT) as a function of the magnetic field at an applied bias of +5 mV between the FM electrodes. Irradiations were performed at RT using the 15 UD Pelletron accelerator at Nuclear Science Centre. Both the undoped and Gd-doped junctions were irradiated with 70 MeV Si at a fluence of 5×10 11 ions/cm 2 and with 200 MeV Ag at a fluence of 1×10 11 ions/cm 2 . Thus, the fluences correspond to typically 10 7–10 8 ion “hits” for the 10−4 cm 2 devices. In this article, we present the data corresponding to the above two representative fluences chosen in a way such that the defect overlap remains incomplete to indicate the damage efficiency of these ions. For both ion-beam energies used here, the electronic energy loss, S e , values in all the junction layers are 2–3 orders of magnitude higher compared to the nuclear energy loss, S n , and therefore, the expected changes of the MTJs can be mostly attributed to the electronic excitation. The S e and S n values in different layers have been listed in Table 1. In general, 200 MeV Ag ions deposit about four times higher energy in the junction layers as compared to the 70 MeV Si ions. The penetration range of Si and Ag ions are much greater (10 μm) than the total film thickness (27 nm) of the devices, as given by the SRIM 2003 code [20]. The samples were mounted on a copper target holder with thermally conducting silver paste. The ion flux was kept low (≈10 9 ions/cm 2 s) to prevent sample heating during irradiation. Such flux value is comparable to extreme industrial environments but greater by a factor of 10 2–10 3 to that encountered in space during solar flares [18]. All the samples were irradiated in a cryopumped vacuum chamber (under a vacuum of 1×10 7 mbar) using a 1 cm × 1 cm uniformly scanned beam in the secondary-electron-suppressed geometry.

3 Results and discussion

The magnetoresistance curves at RT for MTJ-1 and MTJ-2 junctions before and after irradiation have been shown in Figures 1 and 2, respectively. The pristine undoped junction shows a TMR of 20.3% (Fig. 1a), which drops to 18.2% upon Si irradiation (Fig. 1b). From Figure 2, it is observed that the pristine Gd-doped junction shows a TMR of 18.3% (Fig. 2a), which after Si irradiation drops drastically to 0.3% (Fig. 2b). In both cases, however, irradiating with Ag ions leads to a complete destruction of the junctions (Figs. 1c, 2c).

In order to understand the role of Si irradiation on the observed changes in the magnetic properties of the MTJs, let us invoke the appropriate models, which deal with the interaction of SHI with matter. At present, two theoretical models, Coulomb explosion [21] and thermal spike model [22] are used to explain local excitation of the lattice by energy transfer from the highly excited electronic system to the lattice atoms. Both mechanisms can result in atomic transport to modify the interfaces of a SHI irradiated layered structure having widely different kind of materials. However, such modifications are not straightforward, but depend a lot on the material properties, deposited energy density, and the ion species. In most cases, materials’ modification by SHI irradiation exhibits a threshold behavior in terms of S e , beyond which the defect production efficiency increases to a great extent.

It is known that the threshold S e value for Co is 30–40 keV/nm [17]. Therefore, Co layer is expected to be heavily damaged at 200 MeV Ag ion (S e = 36 keV/nm) irradiation, whereas it should not be affected much by Si

<table>
<thead>
<tr>
<th>Ion</th>
<th>Layer</th>
<th>S e  (keV/nm)</th>
<th>S n  (keV/nm)</th>
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</thead>
<tbody>
<tr>
<td>Si</td>
<td>Ni 80 Fe 20</td>
<td>7.93</td>
<td>8.42×10 3</td>
</tr>
<tr>
<td>Al 2 O 3</td>
<td></td>
<td>2.49</td>
<td>2.18×10 3</td>
</tr>
<tr>
<td>Co</td>
<td></td>
<td>7.90</td>
<td>8.06×10 3</td>
</tr>
<tr>
<td>Gd</td>
<td></td>
<td>4.73</td>
<td>5.03×10 3</td>
</tr>
<tr>
<td>Ag</td>
<td>Ni 80 Fe 20</td>
<td>35.47</td>
<td>1.09×10 1</td>
</tr>
<tr>
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<td>11.42</td>
<td>2.74×10 2</td>
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<tr>
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</tr>
<tr>
<td>Gd</td>
<td></td>
<td>20.88</td>
<td>6.80×10 2</td>
</tr>
</tbody>
</table>
ions ($S_e$ 8 keV/nm). To the best of our knowledge, there is no experimental study of SHI irradiation on the other FM layer, Ni$_{80}$Fe$_{20}$. On the other hand, although it is well known that bulk Al$_2$O$_3$ is one of the good radiation-resistant insulators [23] and is used in strong radiation environment, its response to SHI irradiation in the form of an ultrathin film could be different. In fact, different behaviors in case of bulk and thin film form has been observed in case of SHI irradiation of other materials like Ge [24,25]. Nevertheless, creation of atomic displacements and/or their clustering in individual layers and across the interface is expected to give rise to both strong spin dependent and spin independent scattering accompanied by a concomitant decrease in the TMR values.

In the above context, our data in Figures 1a and 1b show a reduction in the TMR by 2.1% after irradiation with Si ions. It may be mentioned here that in a recent report on irradiation of MTJs with Al$_2$O$_3$ barrier [18], similar reduction in TMR value has been observed. They attribute it to limited interface mixing at the FM/I interface or oxygen depletion from the Al$_2$O$_3$ barrier, which are spatially localized. Such localized damage zones caused by SHI induced defects has been modeled as a quantum point contact (QPC) [26] and according to this model, and other tunneling theories of Simmons [27] and Brinkman, [28] the amount of current that tunnels through an energy barrier is exponentially proportional to the barrier thickness and its height. A similar situation can also be envisaged here where SHI induced defect formation in the Al$_2$O$_3$ layer could be approximated by the formation of multiple conducting paths in parallel (due to defect states in the barrier potential) and thus decreasing the overall junction resistance. This could be followed by a small degree of atomic redistribution across the FM/I interfaces as the ions pass through the layers.

A further look at the TMR plots in Figure 1 reveals that for undoped and pristine junctions the switching field for Ni-Fe is 6 Oe and that for Co is 17.5 Oe, which changes to 9.1 and 10.6 Oe, respectively. Such changes occurring in the switching field is probably due to SHI induced softening of the ferromagnetic Co layer by making it a fine-grained FM or smoothening the interface with the barrier layer. It has been observed for Co thin film systems [12, 29] that irradiation induced defects and modification in the film properties as grain size and strain leads to a decrease in the coercivity. At the same time the Ni$_{80}$Fe$_{20}$ layer becomes magnetically harder through the creation of defects and their clusters, which may act as domain pinning centres. Enhancement in coercivity in antiferromagnet/ferromagnet layers (AF/F) like NiFe/FeMn, MnF$_2$/Fe have been observed and ascribed to the magnetic frustration in the AF/F interface causing local energy minima, which effectively pin the propagating domain walls in the FM [14,30].

On the other hand, as mentioned earlier, it is observed that TMR value of the Gd-doped junctions (MTJ-2)
drastically decreases to 0.3% followed by a huge drop (factor of 10\(^3\)) in the tunnel resistance. This is accompanied by the switching field values changing from 8.9 Oe and 10.6 Oe for Ni-Fe and Co to 8.7 Oe and 12.9 Oe, respectively. Such a large change in the TMR clearly indicates that the introduction of Gd in the barrier layer makes it extremely sensitive to Si ion irradiation under similar conditions. In order to understand the behavior of the junction resistance, detailed SRIM 2003 simulation was performed for a layer configuration of MTJ-2. This shows that as a result of Si irradiation, Gd dopant atoms are pushed away from the barrier relatively more towards the Co bottom layer. Therefore, in addition to our above explanation related to MTJ-1, one might expect intermixing at the FM/I interfaces which acts as additional source of spin dependent scattering. This would result into a significant change in the properties of the interfaces and in turn deteriorate the barrier properties.

For the 200 MeV Ag irradiation, even at a lower fluence of \(1 \times 10^{11}\) ions/cm\(^2\), Si induced damage cross-section increases substantially. For both types of junctions, Ag ion is expected to cause heavy damage mainly through the modification of Co and the Al\(_2\)O\(_3\) barrier layer. In particular, SRIM simulation for the Gd-doped MTJ shows large atomic rearrangements to occur due to recoil implantation of different species constituting the FM/I/FM layers. This leads to the complete destruction of the tunneling characteristics.

4 Conclusion

In summary, the influence of SHI irradiation on the junction characteristics of doped and undoped FM-I-FM tunnel junctions has been studied. A small (2.1%) but irreversible decrease in TMR value is observed due to Si irradiation for the undoped case, whereas it shows a dramatic decrease for Gd-doped junctions. A complete destruction of the MTJs in both cases is observed by Ag irradiation. The results are attributed to the modification of interface as well as different layers caused by SHI irradiation. Further studies are underway to correlate these properties with structural changes occurring in the tunnel junctions including temperature and annealing effects under similar SHI irradiation.

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