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Swift heavy ion induced interface modification in metal/Ge systems

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Abstract : This paper reports on swift heavy ion induced interface modification in metal/Ge thin film bilayer systems. 100 MeV Au^{7+} ions have been used for irradiation of the UHV grown thin films of Co/Ge and Ni/Ge at three different temperatures of 100 K, 300 K and 385 K and with fluences in the range of 5×10^{12} ions/cm^2 to 1×10^{14} ions/cm^2. Irradiation induced interfacial changes were studied by Rutherford backscattering spectrometry. The changes in the surface morphology and other structural aspects have been studied by transmission electron microscopy. Interface modification has been explained in terms of synergetic effect of electronic and nuclear energy loss processes.

Keywords : Swift heavy ion irradiation, alloy formation, electron microscopy.

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1. Introduction

Irradiation of solids with swift heavy ions (SHI) results in structural modification as well as alteration of phases. Ion beam mixing (using ions of a few hundred keV energy) of thin metal/semiconductor systems has been widely used to form both equilibrium and metastable phases. In addition to lower processing temperature, this technique offers a high degree of spatial selectivity. However, ion beam mixing using SHI remains mostly an unexplored area of research [1–4]. The important point in SHI induced materials modification is the fact that the energy loss process is dominated by high electronic excitation unlike the keV ion beam mixing process, which is mostly governed by nuclear energy loss. In most cases, materials modification by SHI irradiation exhibits a threshold behaviour in terms of electronic energy loss, S_e, beyond which the defect production efficiency increases to a great extent [5–7]. Due to energy confinement, the response to SHI irradiation for thin films could be quite different as compared to the bulk [8]. In case of layered structures, such energy deposition may lead to atomic intermixing across the interfaces (ion beam mixing).

In this paper, we report on SHI-induced interface modification in Ni/Ge and Co/Ge thin film systems under 100 MeV Au ion irradiation at 100 K, 300 K and 385 K. Rutherford backscattering spectrometry (RBS) and cross-sectional transmission electron microscopy (XTEM) have been employed to study the irradiation induced changes across the interfaces.

2. Experimental details

Electron-beam evaporation technique was used to grow the Ni/Ge and Co/Ge bilayer samples (with Ge as the top layer in both the cases) on ultrasonically cleaned Si substrates in a UHV chamber with the base vacuum of 2×10^{-9} mbar. The bilayer structures were uniformly irradiated at 100 K, 300 K and 385 K using 100 MeV Au^{7+} ions with fluences between 5×10^{12} and 1×10^{14} ions/cm^2. The range of the 100 MeV Au ions and the energy deposited by them were calculated by SRIM-2003 code [9]. Accordingly, the implanted species would

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penetrate deep (~5 μm) into the Si substrate. The electronic and nuclear energy losses (S_e and S_n, respectively) for 100 MeV Au ions in Ge are 16.88 keV/nm and 0.43 keV/nm, in Ni are 31.64 keV/nm and 0.83 keV/nm and in Co are 33.32 keV/nm and 0.77 keV/nm, respectively. Irradiation induced changes across the Ni/Ge and Co/Ge interfaces were studied by RBS with 1.6 MeV He^+ ions, while changes in the microstructure of the films before and after SHI irradiation were studied by XTEM with 200 keV electrons.

3. Results

(A) Co/Ge system :

Figure 1 presents the RBS spectra of Co/Ge samples before and after irradiation by 100 MeV Au ions at different temperatures using an ion fluence of 1×10^{14} ions/cm^2. Comparing these spectra, one observes an upward shift in the RBS signal from the base line (at the Co/Ge interface) and a peak broadening of Ge corresponding to irradiation temperatures of 300 K and 385 K. These aspects could be arising due to interface mixing and/or surface non-homogeneity under irradiations at 300 K and 385 K, while it is insignificant in case of 100 K.

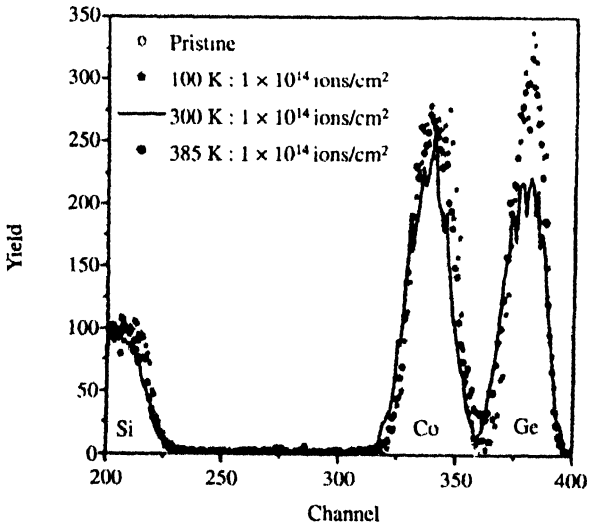

Figure 1. RBS spectra obtained from Co/Ge bilayer before and after 100 MeV Au irradiation at different temperatures.

From the XTEM of the pristine sample (image not shown), it is observed that the Ge layer (55 nm) is amorphous, while the underlying Co layer (49 nm) is polycrystalline in nature. After 300 K irradiation, the Ge layer gets severely modified (Figure 2(a)). Magnified image (not shown here) shows a thin intermixed layer of Co-Ge to get formed across the Co/Ge interface, which is consistent with our RBS results. The corresponding high-resolution lattice images presented in Figure 2(b)

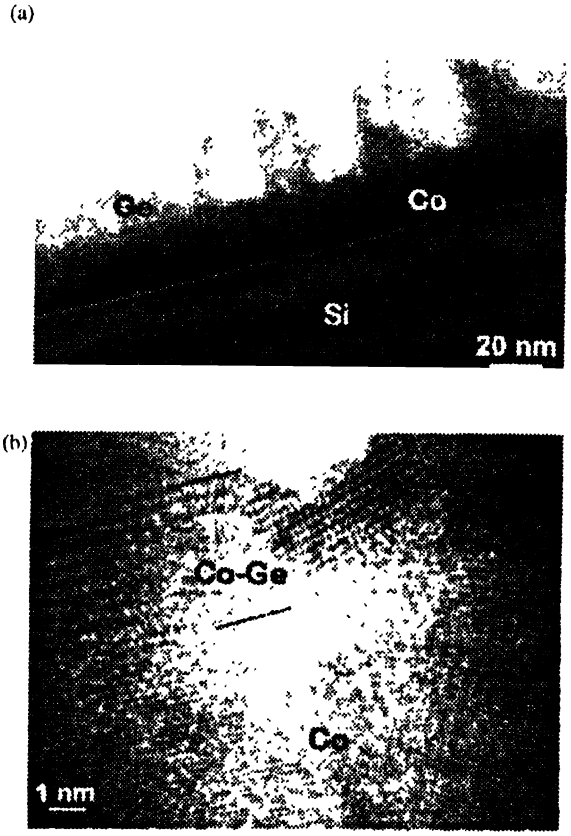

Figure 2. Cross sectional TEM bright field images obtained from Co/Ge bilayer irradiated by 100 MeV Au ions (at a fluence of 1×10^{14} ions/cm^2) at 300 K : (a) low-magnification image showing all the layers, (b) high resolution lattice images from the mixed Co-Ge layer and the underlying Co layer.

indicates the Ge layer to remain as amorphous, while the newly formed mixed Co-Ge layer (~3–4 nm) is polycrystalline, having a different d-spacings compared to the underlying Co layer. It can be mentioned here that three equilibrium phases of the Co-Ge system have been predicted in the phase diagram, *viz.* Co_2Ge, CoGe and $CoGe_2$ [10]. Unfortunately, X-ray diffraction data are not available for the Co_2Ge phase and hence the calculated d-spacings could not be compared with the same. Therefore, it is difficult to infer unambiguously about the phase formed in the present case. However, from the point of the effective heat of formation [11], the Co_2Ge phase has the highest value among the three phases and accordingly should form first. On the other hand, for 100 K irradiation, small surface cratering takes place while for 385 K irradiation, modification caused to the Ge

layer is much less compared to the case of 300 K and small void like structures get formed into it. However, the peak broadening and the upward shift from the base line in the corresponding RBS spectrum of the 385 K irradiated sample could be occurring due to interface mixing. Detailed XTEM studies are underway to confirm this aspect.

(B) Ni/Ge system :

Like Co/Ge, here also irradiation at 300 K brings into maximum changes at a fluence of 1×10^{14} ions/cm^2. In a recent paper [4], we have shown that irradiation of Ni/Ge performed at 300 K leads to the formation of a uniform layer (~6–7 nm) of Ni_2Ge (at the Ni/Ge interface) whereas 100 K irradiation does not lead to any significant mixing. Therefore, here we present only the results of irradiation performed at 385 K. Figure 3 shows the RBS

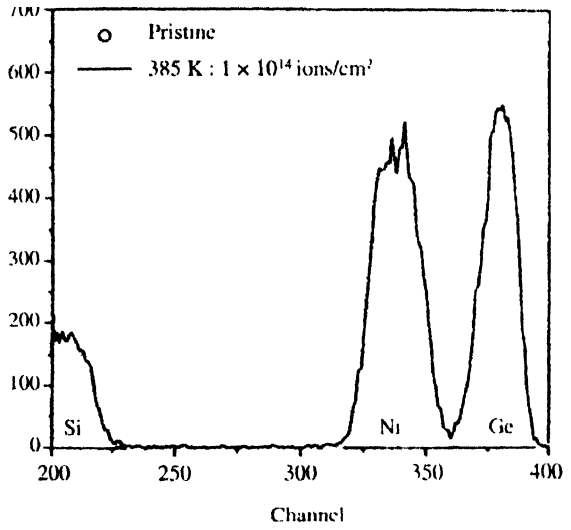

Figure 3. RBS spectra obtained from Ni/Ge bilayer before and after 100 MeV Au irradiation at 385 K.

spectra obtained for Ni/Ge samples before and after 100 MeV Au irradiation at 385 K at a fluence of 1×10^{14} ions/cm^2. Comparing these two spectra, it is observed that a small reduction in the Ge signal takes place followed by a small amount of upward shift in the signal near the interface. This could arise due to irradiation induced surface inhomogeneity and/or intermixing.

We carried out XTEM analyses on these samples to determine their thickness and microstructure. The pristine sample shows a layer structure of 46.5 nm Ge and 38 nm Ni on Si substrate. High-resolution TEM images indicate that the top layer of Ge is amorphous, while the Ni layer underneath is a polycrystalline one [4]. The other related features of 100 MeV Au irradiated Ni/Ge bilayers irradiated at 100 K and 300 K have been discussed in Ref. [5]. Figure 4 shows the XTEM image

Figure 4. Low magnification crosssectional TEM bright field image obtained from Ni/Ge bilayer irradiated by 100 MeV Au ions (with a fluence of 1×10^{14} ions/cm^2) at 385 K.

taken after irradiation of Ni/Ge sample at 385 K. It is clear that the Ge surface becomes quite uneven followed by the formation of void structures in it. However, high-resolution lattice imaging shows the formation of a thin Ni-Ge alloy layer in between the Ni and Ge layers. These explain the nature of the RBS spectrum in Figure 3 corresponding to Au irradiation at 385 K.

4. Discussion

Let us explain our observations and look for the probable cause of the observed Co-Ge and Ni-Ge phase formation in terms of interaction of SHI with matter. As SHI passes through a material, it leads to production of defects and formation of a latent track depending on the sensitivity of the material and the deposited energy density. Once S_e crosses a certain threshold value, formation of ion track sets in as a result of local melting and subsequent quenching [6,12]. Recently, SHI mixing in Ni/Si, (where both the materials have undergone subthreshold irradiation) has been shown [1] and a solid state reaction has been proposed to occur in the hot zone around the ion path.

In our case, since the S_e values are ~40–50 times larger than the respective S_n values. Since corresponding to these Sn values, Ar induced mixing in Ni/Ge and Co/Ge systems has earlier been reported [13–15], the contribution of S_n cannot be neglected in our case. For 100 MeV Au irradiation of Ni/Ge and Co/Ge bilayer systems, the S_e value for Ge is 3.2 times higher than its

threshold and therefore, it is expected that Ge will get severely modified due to S_e [16]. On the other hand, S_e value for bulk Ni is 2.1 times lesser than the reported S_e value up to which Ni to remain unaffected [17]. However, Sn induced point defects are still created in Ni (~1.4/ion/Å from detailed SRIM simulation). These point defects would be expected to be highly mobile at room temperature. Similarly, the threshold S_e value for the bulk Co is known to be ~30–40 keV/nm [5], which is comparable to the present case. A good amount of point defect creation in Co (~1.0/ion/Å from detailed SRIM simulation) would be expected here also. S_n-induced effects being cumulative, a large number of point defects would be available to both Co and Ni near the interface. Therefore, response of Ni and Co would be expected to be quite different as compared to the case where $S_n << S_e$. This is followed by the neighouring Ge layer to undergo a lot of atomic redistribution caused due to SHI induced mass transport [4], which is common for only amorphous materials [18]. Thus, in the presence of a large number of point defects created by S_n, S_e-induced effects (as described earlier) would lead to atomic movements across the *highly* reactive Ni-Ge and Co-Ge interfaces to cause interface mixing. For 100 K irradiations, less atomic mobility leads to almost negligible mixing, whereas for the case of 385 K the void structure seen in Ge could be the result of a high density of defect clusters. The details of such temperature dependent morphological changes in Ge as a result of SHI irradiation will be published elsewhere.

5. Summary

In this work, we have shown the interface modification in Metal/Ge (Metal = Co and Ni) systems under 100 MeV Au ions at different temperatures, *viz.* 100 K, 300 K and 385 K. SHI irradiation at 300 K leads to significant interface mixing followed by morphological changes in Ge. The results have been explained in terms of synergetic effect of nuclear and electronic energy loss.

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