Study of interface electronic properties of annealed Fe/Al multilayers using synchrotron radiation

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1. Introduction

Transition metal (TM) aluminides are a well-known class of structural intermetallics that possesses a unique combination of several desirable properties such as high melting point, oxidation and corrosion resistance. In particular, iron-aluminium alloys constitute one of the most interesting systems to investigate the effect of size, composition, order, and temperature on the magnetic properties [1]. It is known that in FeAl alloy system there exist mainly two ordered FeAl phases, namely Fe_3Al having DO3 structure and is ferromagnetic, whereas, FeAl phase having CsCl lattice structure and is non-magnetic in nature. It is found that starting from pure Fe, the gradual substitution of the Fe sites by Al leads to a progressive decrease of the saturation magnetization and the alloy becomes nonmagnetic around 35% Al [1]. Similarly, resistivity initially increases with Al concentration and shows a maximum at 35% Al and then decreases with further increasing Al concentration [2]. All these properties are basically dictated by the interactions between the Al-sp and TM-d bands. The subtle changes in the p-d hybridisation brought about by a variation in Al concentration affect the chemical bonding and phase stability of intermetallic alloy. Therefore, in order to understand the reason behind this anomaly, we have investigated these annealed Fe/Al MLS using GIXRD and valence band photoelectron spectroscopy techniques.

2. Experimental

The multilayer samples [Fe (70 Å)/Al (42 Å)]$_{x15}$ (sample A) and Fe (50 Å)/Al (70 Å)]$_{x15}$ (sample B) used in the present case were deposited on float glass using electron beam evaporation technique under the UHV conditions in order to ensure an average 3:1 and 1:1 atomic composition of the deposited multilayer samples. The deposited MLS are annealed at 400°C for 4hr in a vacuum better than $1x10^{-7}$ Torr. For the determination of micro-structural changes in the MLS upon annealing, grazing incidence X-ray diffraction (GIXRD) technique is used. The valence band photoemission measurements (VBPE) were carried out on TGM beamline [3] installed at Indus-1 using synchrotron radiation. The measurements were carried out in a base pressure of $5x10^{-10}$ Torr. For photon energy (hν) of 134eV, the overall energy resolution, including the analyser, was estimated to be better than 0.65eV.

3. Results and discussion

The GIXRD measurements (not shown here) reveal the formation of Fe_3Al and FeAl intermetallic phases for the MLS annealed at 400°C/4hr having atomic concentration ratio of 3:1 and 1:1, respectively.

Further in order to understand the origin behind the anomaly in resistivity and magnetic behaviour with increasing Al concentration, we have carried out the valence band photoemission measurements on sample A and B. As we have already mentioned that all these properties are basically dictated by the interactions between the Al-sp and TM-d bands and the VB photoemission spectra directly relates to the density of occupied electronic states near the fermi level. Fig. 1 shows the valence band photoemission spectra of corresponding Fe/Al multilayer samples recorded after 25 minutes of sputtering, where the contributions of both Fe and Al are seen simultaneously. VB photoemission measurements carried out on sample A mainly show the following three distinct features: (i) broad Fe-3d photo-emission bands around 0.56, 1.82 and 2.98 eV below the fermi level (ii) O-2p band at 5.66 eV and (iii) Al-Ox band at 7.72 eV due to the hybridisation of π and σ molecular orbitals of Al-3sp and O-2p along with Al-3s band at around 10.40 eV. The observed features of Fe-3d, O-2p and Al-3s bands are in excellent agreement with results obtained by A. Arranz et al. [4]. However, in case of sample B, the density of states of Fe and Al near to Fermi level is modified significantly i.e, becomes broader and flattens as compared to sample A. The binding energy values of Fe-3d band are shifted towards lower value i.e. at 0.42, 1.6 and 2.8eV, respectively, with corresponding increase in Fe-3d and decrease in Al-3s density of states as can be seen from fig. 1, suggesting the charge transfer from valence Al-3p and Al-3s electrons to minority 3d orbitals of Fe due to the strong hybridisation of sp-d states at fermi level giving rise to Fe-Al metallic like properties. In order to show the modified density of states of Fe-3d near to fermi level, they are plotted together in the inset of fig. 1.

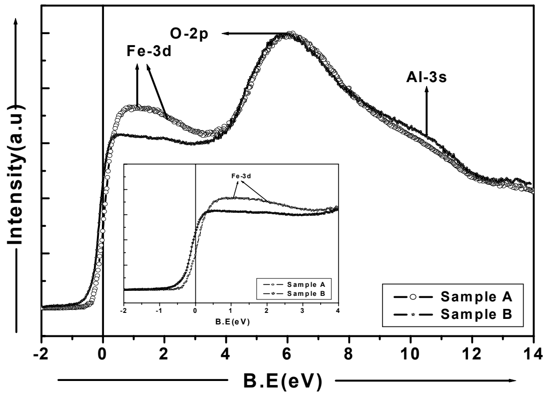

Fig.1 Valence band spectra of (A) [Fe (70 Å)/Al (42 Å)]$_{x15}$ and (B) Fe (50 Å)/Al (70 Å)]$_{x15}$ MLS annealed at 400°C for 4hr.

One can easily understand the above-observed changes in the valence band behaviour upon increasing Al concentration from the electronic structure of Fe and Al atoms. The outermost valence orbitals of Fe and Al atoms are ($3d^6 4s^2$) and ($3s^2 3p^1$), respectively. Due to unfilled 3d orbital of Fe and 3p orbital of Al, the orbitals are split into spin ↑ and spin ↓ states. However, the 3s orbital of Al are very close in energy to the 3d ↑ orbital of Fe, providing the grounds for a strong overlap between Fe-3d and Al-3s as these electrons are alloyed. More interestingly, the 3p electrons of Al lie at a higher energy than the 3d and 4s electrons of Fe. Thus, as these elements are alloyed, one would expect a charge transfer from Al-3p to Fe-3d, as the 4s orbitals of Fe are full. Therefore, the charge transfer would have an immediate effect on the resistivity and magnetic moment of Fe due to filling of the 3d orbitals.

Conclusion

Valence band photoemission measurements show that the changes in the electronic structure brought about by the hybridisation of the Fe-3d and Al-3sp due to the charge transfer from Al to Fe is found to be responsible for the observed anomaly in the electrical resistivity and magnetic behaviour.

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References

1. H. Sato et al., Phys. Rev. 114 (1959) 1427.
2. B. V. Reddy et al. Intermetallics 8 (2000) 1197.
3. S. M. Chaudhari et al., Curr. Sci. 82 (2002) 305.
4. A. Arranz et al., Surf. Sci. 521 (2002) 77.

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