

## Study of self-diffusion of iron in amorphous Fe-Zr alloys using nuclear resonance reflectivity

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Mössbauer spectroscopy has been used conventionally to study atomic diffusion through the changes in the spectral line shape [1]. The range of diffusivity accessible through this type of technique depends upon the lifetime of the Mössbauer excited state and is typically in the range  $10^{12}$  to  $10^{13}$  m<sup>2</sup>/sec. More recently a much more powerful method of nuclear resonant forward scattering of synchrotron radiation has been used which relies on the time domain measurement of the scattered intensity [2]. The present experiment is aimed at using nuclear resonance reflectivity from isotopic multilayers [3] to study self-diffusion of iron in some amorphous and nanocrystalline alloys having diffusivities several order of magnitude smaller than those accessible using the above concepts. The systems studied are amorphous FeZr and nanocrystalline FeNzr alloys. Isotopic multilayers of the systems being studied were prepared having structure Substrate/[<sup>57</sup>FeZr(30Å)/FeZr(40Å)]×10 and Substrate/[FeZrN(40Å)/<sup>57</sup>FeZrN (30Å)]×20. At nuclear resonance energy the reflectivity for the multilayer exhibits Bragg peak due to isotopic periodicity. Diffusion of iron in the multilayer (induced by thermal annealing) would result in a decrease in the contrast between the two layers and thus a decrease in the height of the Bragg peak. The diffusion length (L) can be obtained using the relation

$$\ln [I(t)/I(0)] = 2 \pi^2 n^2 L^2 / d^2$$

Where I(t) is the intensity of the nth order Bragg peak after annealing for time t, L is the diffusion length and d is the bilayer periodicity. The nuclear reflectivity is separated from the prompt reflectivity of the non-resonant photons by filtering in time domain.

Figure 1 gives the nuclear reflectivity of FeZr multilayer after isochronal annealing at different temperatures. The height of the Bragg peak shows monotonous decrease with increasing annealing temperature, indicating interdiffusion of <sup>57</sup>Fe and <sup>56</sup>Fe. Intensity of the Bragg peak was obtained by first removing the small angle scattering background by multiplying the intensity with a factor q<sup>4</sup>. The diffusivities obtained at 423K and 455K are  $D_{423} = 4.83 \times 10^{-23}$  m<sup>2</sup>/s,  $D_{455} = 8.7 \times 10^{-23}$  m<sup>2</sup>/s, which are comparable to the values for similar alloys [3].

A detailed study of the kinetics of self-diffusion of Fe was done in the nanocrystalline sample FeNzr using isothermal annealing at three different temperatures. Figure 2 gives the annealing time dependence of  $\ln[I(t)/I(0)]$  at 393K. The slope of the curve initially decreases at a faster rate and then becomes constant beyond an annealing time of 16 minutes. The initial faster change in the slope may be associated with the structural relaxation in the system. Slope of the later part

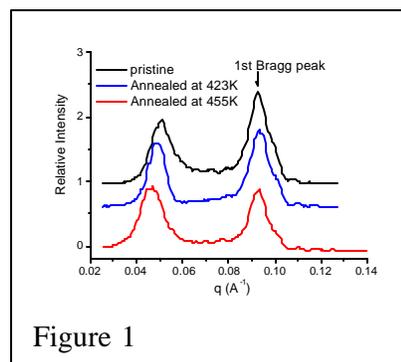


Figure 1

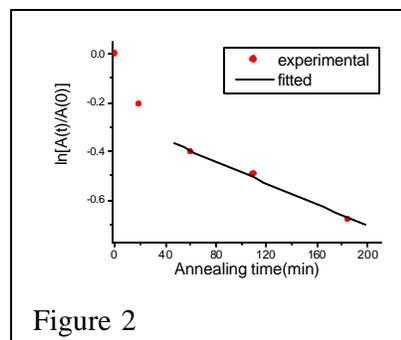


Figure 2

of the curve is used for determining the diffusivity in the structurally relaxed state. Taking  $D = D_0 \exp(-E/k_B T)$ , measurements at three different temperatures 393K, 423K and 448K yield  $D_0 = (3.5 \pm 0.8) \times 10^{-13} \text{ m}^2/\text{sec}$ ,  $E = (0.80 \pm 0.01) \text{ eV}$  (Fig. 3).

It is interesting to note that the measurements provide a clear indication of the structural relaxation of the system during the initial stages of annealing. **Typical diffusion lengths in this region are 2Å to 10Å.** Such a small diffusion length is not accessible through the conventional tracer techniques [3], as the typical depth resolution of these techniques is a few tens of Angstrom. It is planned to do more detailed study during the initial stages of annealing in order to study the kinetics of structural relaxation in the system.

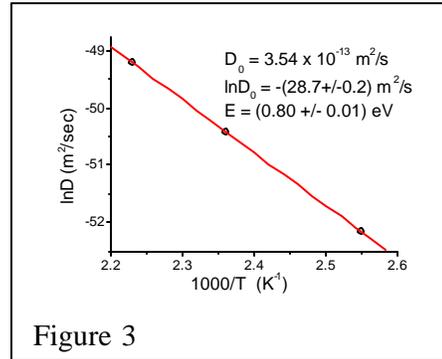


Figure 3

Depth profiling of a single 50Å thick FeZr marker layer was also done using x-ray standing waves in a multilayer having structure: Substrate/ Pt (700Å)/FeZr (175Å)/<sup>57</sup>FeZr (50Å)/FeZr (175Å). Total reflection of x-rays from Platinum layer results in formation of x-ray standing wave. With increasing angle of incident, the antinodes of the standing wave field scan through the <sup>57</sup>FeZr marker layer [4], resulting in a modulation of the nuclear resonance fluorescence from <sup>57</sup>Fe nuclear. Such modulations are observed only in the delayed fluorescence and delayed forward scattering.

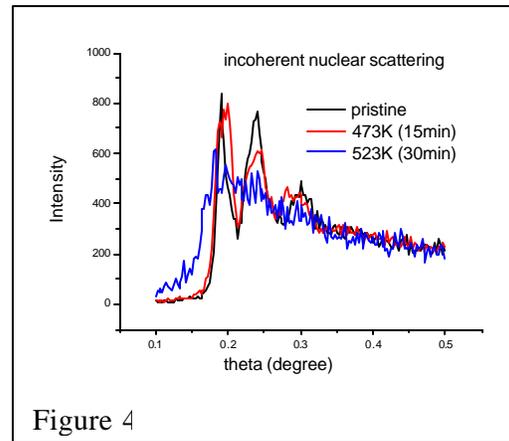


Figure 4

Fig.4 gives the incoherently scattered x-rays from the above multilayer after different stages of annealing. The fluorescence peaks get broadened with increasing annealing and after annealing at 523K for 30 min. peaks have totally disappeared indicating that <sup>57</sup>Fe has been distributed almost homogeneously inside the film. A detailed analysis of the experimental data using Parratt's formalism is being done in order to extract the depth profiles of <sup>57</sup>Fe marker layer. These measurements clearly show that nuclear resonance fluorescence under standing wave conditions can be used for depth profiling of isotopic marker layers. Thus, self-diffusion measurements can also be done on a single <sup>57</sup>Fe marker layer.

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