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Fe^+ ion irradiation induced changes in structural and magnetic properties of iron films

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ABSTRACT

490 keV Fe^+ ion irradiation of 200 nm thick Fe films was found to induce both structural and magnetic changes. Both, the lattice constant and the grain size increase as a function of dose and both properties follow the same power law. Irradiation induces a depth dependent magnetic profile consisting of two sublayers. The top Fe sublayer has a magnetic moment higher than that of the Fe before the irradiation whereas the bottom sublayer lower. The two sublayers are connected with the effects of Fe^+ irradiation, i.e. the top sublayer with the depth in which mainly radiation damage occurs whereas the bottom one with the implantation of impinging Fe^+ ions. The magnetic moments of the two sublayers have a non-monotonous variation with irradiation dose depicting a maximum for the top sublayer and a minimum for the bottom one at 96.2 dpa ('displacements per atom'). The magnetic moment enhancement/reduction is discussed in relation with the atomic volume variation in the case of atom displacements and/or implantation effects.

1. Introduction

Although significant progress has been made in the development of improved radiation resistant alloys, the so far developed alloys are unable to withstand the severe operational conditions expected in fusion power plants [1–3]. Therefore, there is need for developing new radiation resistant alloys and to that end there is an international effort aspiring into understanding the effects of neutron radiation damage. In order to materialize this objective multiscale modeling, experimental validation and irradiation campaigns in fission reactors [4,5] are implemented. The last activity suffers from a vital inadequacy concerning fusion applications as the validity of extrapolating the fission neutron irradiation results to fusion environment is questionable. This contention arises from the fact that the energy spectrum of fission neutrons (mean energy of about 2 MeV) within a material is very different to that produced from the 14 MeV fusion neutrons. This shortcoming of the experimental basis of the research on neutron radiation damage to be generated in a fusion reactor can be surmounted by appreciating that the dominant damage arises from Primary Knock-on Atoms (PKA). Therefore, the main features of the neutron induced radiation damage in the materials can be studied by employing self-ion irradiations [6]. As the energy of the impinging ions is well determined averaging effects of different PKA energies can be avoided and also a wide choice of ion energies and fluxes are available for a methodical study of the damage. Implementation of ion irradiation has the additional advantage that the radiation damage arising solely from PKA can be studied by avoiding neutron transmutations e.g. helium and hydrogen production. Further, the ion irradiated samples are not radio-active and they can be examined immediately after irradiations employing all the available material science techniques. Therefore, the experimental results on the damage produced by energetic ions provide significant and comprehensible information on radiation damage and they consist a rewarding testing bed for theoretical models and simulations. It is worth mentioning that the cost of ion irradiation in accelerators is very low in comparison to neutron irradiations in fission reactors.

It was chosen to utilize the above expressed viewpoint on Fe as ferritic/martensitic FeCr based steels are candidate structural materials for the future fusion reactor. The energy of the Fe ions for

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the irradiations was defined to 490 keV as this is the mean energy of the Fe PKA produced by the 14 MeV fusion neutrons impinging on iron based alloys. As the range of this energy ions is of the order of few hundreds of nanometers samples in the form of films have to be employed. Thus, films of 200 nm thickness were employed in order to observe effects arising from both radiation damage and ion implantation. The current works refers to the effect of Fe^+ ion irradiation on the structural and magnetic properties of the 200 nm thick iron films whereas in a previous work [7] 60 nm thick Fe films, in which only radiation damage occurs, were investigated. Irradiations with 490 keV Fe^+ ion beam were performed for different doses. The structural characterization of the samples before and after the irradiations was performed using grazing and normal incidence X-ray diffraction and X-ray reflectivity measurements. The induced magnetic state changes were determined by magnetic hysteresis loop and polarized neutron reflectivity measurements. Magnetic hysteresis loop measurements refer to the magnetic properties of the total film thickness, whereas by polarized neutron reflectivity (PNR) the magnetic moment per atom versus depth is determined.

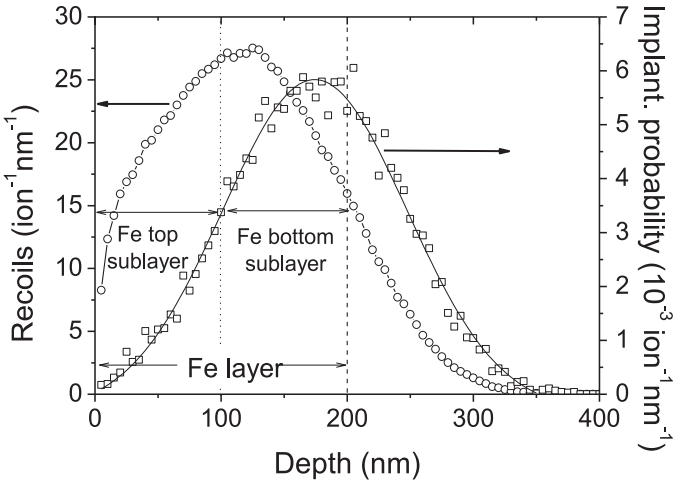

Fig. 1. Recoils and implantation profile for 490 keV Fe^+ on Fe target according to SRIM calculations. For the definition of top and bottom Fe sublayer refer to Fig. 8a.

2. Materials and methods

Iron films having a thickness of 200 nm were fabricated on one side polished (001) silicon wafer substrates using DC magnetron sputtering. A Cr cover layer of 4 nm nominal thickness was deposited on top of the Fe layer in order to prevent oxidization.

Irradiations with 490 keV Fe^+ ion beam were performed at JANNuS facility at CEA-Saclay using an ion flux of around 2×10^{12} ions/(cm^2 s). The samples during the irradiation were placed on a liquid nitrogen cooled flange compensating the heating induced by the beam and thus keeping the sample temperature at 25 °C. The damage is characterized in terms of the average number of times that an individual atom is displaced from its lattice site i.e. 'displacements per atom' (dpa) and this unit is employed throughout this paper. The samples were irradiated for different doses which correspond to a range from 0.5 to 341 dpa (Table 1).

Simulations using the SRIM-2008 software [8] have been performed of the bombardment of 490 keV Fe^+ ions on an iron target using for the displacement energy the value of 40 eV [ASTM standard, 9]. In Fig. 1 are depicted the number of recoils and the ion implantation probability per incident ion versus the depth within the iron target. It is observed that up to a depth of around 100 nm mainly radiation damage occurs and above 100 nm up both radiation and implantation effects take place.

The films were characterized before and after the irradiation for both the structural and magnetic properties. The structural characterization was carried out by X-ray reflectivity (XRR) and X-ray diffraction (XRD) both at normal and grazing incidence angle. The X-ray measurements were performed on a D8 Advance Bruker diffractometer using Cu-Kα radiation and a parallel beam stemming from a Göbel Mirror. The magnetic state of the samples was determined by Vibrating Sample Magnetometry and Polarized Neutron Reflectivity (PNR) measurements. The PNR measurements were performed at PRISM instrument at Laboratoire Léon-Brillouin, CEA-Saclay. An in-plane external magnetic field of 1.2 T was applied in order to magnetically saturate the samples. The incident neutron wavelength was 0.40 nm and the Q-range varied from 0.05 to 2.3 nm^{-1} (Q is the magnitude of scattering vector, $\boldsymbol{Q} = \boldsymbol{k'} - \boldsymbol{k}$, where $\boldsymbol{k'}$ and $\boldsymbol{k}$ the wave vector of the scattered and incident beam, respectively). In PNR measurements we obtain two reflectivities the one, R^+, corresponding to the spin of incident neutron being parallel to the applied magnetic field (spin up) and the second, R^-, to the spin being down. The spin up and down reflectivities can be calculated from the exact solution of the Schrödinger equation for an assumed model of density and magnetic moment profile versus depth. The parameters characterizing the density model (e.g. thickness, density, magnetic moment per atom) are derived by a least squares fit to the experimental data using the SimulReflec software [10].

Table 1
Irradiation parameters.

Ion flux ($\times 10^{12}$ ions/(cm^2 s))	Irradiation time (min)	Dose ($\times 10^{15}$ ions/cm^2)	Dose (dpa)
2.4	1.5	0.2	0.5
	2.5	0.4	1.1
	29	3.6	9.2
	93	10.8	27.6
	308	37.9	96.2
	984	134.1	341

3. Results

3.1. Structural changes induced by the Fe^+ irradiations

Initially any structural changes induced by the Fe^+ ion irradiation of the Fe films were evaluated. For this purpose two techniques were employed, namely X-ray reflectivity (XRR) and X-ray diffraction (XRD) both at normal and grazing incidence angle. The samples were characterized before and after the irradiations.

The least square fit of the XRR data shows that the Fe layer for doses up to 27.6 dpa has an average atomic density similar to that of bulk Fe (8.5×10^{28} atoms/m^3). For higher doses two Fe sublayers (top and bottom sublayer), with about 100 nm thickness each, were found with different atomic densities. The correspondence of these two sublayers with the radiation damage and implantation induced by the Fe^+ ion irradiations is presented in Fig. 1. The top sublayer corresponds to the depth in which radiation damage is the main effect and the bottom sublayer relates to the depth in which both damage and implantation exist. The density of the top and bottom sublayers reaches the value of 8.9×10^{28} at/m^3 and 9.2×10^{28} at/m^3, respectively, for the maximum dose of 341 dpa.

The XRD measurements at normal and grazing incidence angle (GIXRD) reveal a bcc crystalline structure of the as fabricated Fe films. The crystallinity remains even after the irradiation of 341 dpa, whereas it is observed that as the dose increases the (110) Bragg peak moves to lower angles showing an increase of the lattice constant (Fig. 2). The lattice constant increases with the increase of the dose from 0.2859(7) nm for the un-irradiated sample to 0.2869(3) nm for the maximum dose of 341 dpa. A similar

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