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Report:

Method

To investigate the diffusion mechanism in a near surface layer of an Fe_3Si the Nuclear Resonant Scattering (NRS) in grazing incidence geometry was used. NRS is a well established method to study structural and magnetic properties of materials as well as dynamics on an atomistic scale. This method combines the high brilliance of a third generation synchrotron source and the excellent characteristics of Mössbauer Spectroscopy (MS). The NRS allows to perform diffusion studies in the time domain [1,2]. Synchrotron radiation pulses create a collective coherent nuclear state in the sample. During the scattering process this state is destroyed by the diffusion of atoms. This leads to a faster decay of a scattered intensity and allows to derive details about diffusion. This technique was successfully applied to diffusion studies in bulk samples. A combination of NRS and grazing incidence geometry can be used as a very sensitive tool to investigate thin films or surface-regions. This possibility allows a direct comparison of bulk and near-surface diffusion.

Experiment

The film of Fe_3Si was created by annealing of an Fe/Si multilayer produced by Molecular Beam Epitaxy (MBE) deposition on a MgO substrate. The sample was prepared under UHV conditions. The ratio of deposited iron and silicon atoms in each layer corresponds to the ratio of atoms in the well ordered $D0_3$ structure of Fe_3Si (3:1). The exact stoichiometry given by RBS measurements was 76% of Fe and 24% of Si. The formation of the multilayer structure and the process of annealing were followed by X-ray reflectivity measurement (Fig. 1.). A superstructure peak corresponding to a superlattice constant of about 2 nm is clearly visible in the reflectivity curve (arrow). It disappears at about 400 °C and afterwards the reflectivity consists predominantly of thickness oscillations with one frequency. It means a homogenous Fe_3Si film was created. The total thickness determined from Kiessig beats is 21 nm. The CEMS experiments performed on this film after the synchrotron experiment confirmed the stoichiometry results from RBS and revealed its structure ($Fe_{77}Si_{23}$). We suppose that the resulting sample is a $D0_3$ ordered 21 nm thick polycrystalline film of Fe_3Si.

The temperature of the measurement was calibrated by the magnetic transition (ferromagnetic-paramagnetic) of the film (T_c = 550 °C [3]). The diffusion investigations have been performed about 20 degrees above the Curie temperature of Fe_3Si to avoid a beat pattern in the NRS spectra resulting from magnetic hyperfine interactions [1]. The measurements were done at three different incidence angles (1.7 mrad, 2.47 mrad and 3.7 mrad) and three different temperatures (about 10, 20 and 30 degrees above T_c). The shape of the time response for a paramagnetic layer is defined by the chosen incidence angle and by the diffusion model. Enhanced diffusion results in an accelerated decay of the delayed intensity [4,2]. All NRS spectra have been taken at the ID18 beamline of ESRF operating in 16-bunch timing mode in the usual setup for grazing incidence geometry [5]. The vertical dimension of the beam was 150 μm. The sample was kept in UHV chamber dedicated for grazing incidence investigation.

Preliminary results

The spectra at 570 °C for two incidence angles (1.7 mrad and 2.47 mrad) are shown in Fig. 2. Fits to all the spectra were performed using EFFINO programme written by Spiering et al. [6]. All fits were made with constant thickness of the whole sample (21 nm). For the fit model we have chosen a 18 nm bulk-like layer with two distinct iron sites with different weights according to the diffusion model of Sepiol et al. [4] and a 3 nm cover-layer with enhanced diffusion. This cover layer is necessary to achieve satisfying fits. The Mössbauer spectroscopy measurements of Sepiol et al. [4] show different positions of the absorption lines in bulk (distance 0.16(1) mm/s). This distance disappears at higher temperatures (faster diffusion) due to the relaxation effects [7].

In the case of the 3 nm thick near-surface layer, with faster diffusion, a relaxation effect similar to the QMS measurements yielded into satisfying fit [7]. The calculated diffusion constant in the bulk layer (D= (4.0±1.0 10^{-14} m^2/s) agrees well with QMS results [8,7] and tracer diffusion experiments [9]. The calculation of D in near-surface region gives the value of D=(13.0±1.0) 10^{-14} m^2/s.

First attempts to resolve the atomic jump diffusion mechanism in the near surface layer showed promising results nevertheless to determine clear the mechanism a larger temperature range and the angular dependence of the accelerated decay caused by diffusion have to be investigated.

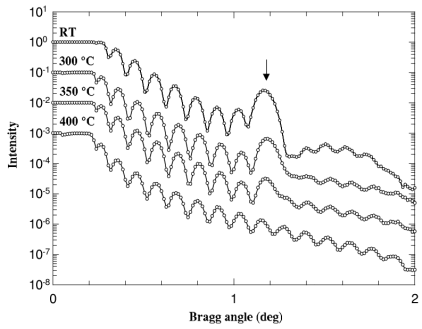

Fig. 1. Annealing process of an Fe/Si multilayer. The magnitudes of curves were shifted on the plots for better visibility

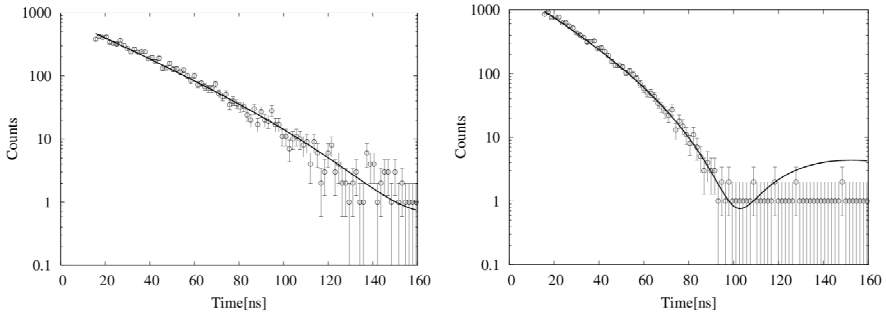

Fig.2.: NRS spectra of a Fe_3Si film on MgO substrate at 570 °C at two diferent incidence angles (1.7 mrad left; 2.47 mrad right). The spectra consist of a contribution from bulk part of the film and a near surface layer with enhanced diffusion. The line is a fit corresponding to the known NN jump diffusion mechanism.

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