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Thermal and irradiation induced interdiffusion in Fe_3O_4/MgO(001) thin film

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ABSTRACT

The interface reactions in an epitaxial 10 nm-thick Fe_3O_4/MgO(001) film were investigated by using Rutherford Backscattering spectrometry (RBS), channeling (RBS-C) and X-ray reflectometry (XRR). The as-grown film had a good crystallinity indicated by the minimum yield and the half-angle value for Fe, respectively, $\chi_{min}(Fe) = 22\%$ and $\psi_{1/2}(Fe) = 0.62°$. Annealing the films under partial argon pressure up to 600 °C led to a large enhancement of Mg out-diffusion into the film forming a wustite-type phase, but the total layer thickness did not change much. Ion irradiation of the film by 1 MeV Ar ion beam caused a strong Fe ion mixing resulting in a large interfacial zone with a thickness of 23 nm.

1. Introduction

Magnetite (Fe_3O_4) – the first known magnetic material has been recently investigated extensively, since it is viewed as a promising candidate for room-temperature spintronic applications [1,2]. It crystallizes in the inverse spinel cubic structure with a lattice constant of $a = 8.396$ Å, 32 oxygen anions O^{-2} form a close-packed face-centered-cubic fcc lattice, eight Fe^{3+} ions locate in the tetrahedrally coordinated A-sites, while 16 octahedrally coordinated B-sites are occupied randomly by eight Fe^{2+} and eight Fe^{3+} ions. A well-known feature of magnetite is a so-called Verwey transition (T_V) around 125 K commonly interpreted as a long-range charge-ordering in the octahedral Fe sublattice. For a review of magnetite properties, see [3,4]. Recently, intensive studies have been focused on the structure and properties of the Fe_3O_4(001) surface. The MgO(001) substrate has been used frequently due to the fact that in the (001)-plane the very small lattice mismatch (0.31%) between the Fe_3O_4 film and the MgO substrate provides favorable conditions for the molecular beam epitaxial (MBE) growth. However, many features of structural, electronic and magnetic properties of these thin film materials are still not fully understood. For a review see [5]. Additionally, large size-effects have been revealed for the epitaxial magnetite thin films. A large reduction of the Verwey temperature and a strong deviation from the bulk properties were observed due to the formation of a magnesium rich phase near the Fe_3O_4/MgO interface [6].

The goal of the experiments, reported here, is to investigate the surface and the interface stoichiometry and crystalline quality of epitaxially-grown Fe_3O_4/MgO(001) films using Rutherford backscattering spectrometry (RBS), channeling experiments (RBS-C) and X-ray reflectometry (XRR) measurements. Details of our study on magnetite films in the as-grown state have been reported elsewhere [7]. In this work we focus to investigate the interface properties influenced by thermal annealing and ion irradiation for the 10 nm-thick Fe_3O_4/MgO(001) film.

2. Experimental details

The film preparation and characterization by low-energy electron diffraction (LEED) and conversion electron Mössbauer spectroscopy (CEMS) have been carried out using a multi-chamber UHV system. After preparation and in situ characterization by LEED, the samples were characterized by CEMS technique using a proportional He/CH_4 flow detector and 200 mCi ^{57}Co (Rh) source.

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Details of sample growth and characterizations by LEED and CEMS have been reported elsewhere [5,6].

The RBS and RBS-C experiments were performed at the Institute of Nuclear Physics of the University Frankfurt/Main [7]. RBS measurements were carried out using a 2 MeV He^+ ion beam at a backscattering angle of 171°. For the data evaluation the computer code SIMNRA [8] was used. The layer thickness values (in nm) were estimated from the simulated RBS areal density values (in at./cm^2) using the bulk density of magnetite taking into account the small change in density related to the Mg diffusion and/or the value for density determined from XRR measurements. Since the film is very thin, the RBS spectra were collected and evaluated at different tilt angles (φ was in the range of 0–45°) in order to improve the depth resolution. RBS-C experiments were performed using 1.6 MeV He^+ ions at a backscattering angle of 160°. X-ray reflectometry (XRR) measurements were performed at a Seifert two-circle diffractometer using a rotating anode with 40 kV and 120 mA, and a LiF monocromator and a slit system for separating the Cu K-α1 line. For XRR data analysis the Seifert Reflectivity software was used.

After the CEMS experiments each sample was cleaved into two pieces (10 mm × 5 mm × 1 mm). One piece was used for diffusion studies. It was annealed at 300° C and then at 600° C, each for 15 min in a partial argon pressure (0.7 atmosphere). The second piece was used for ion beam mixing investigations; it was irradiated by 1 MeV Ar ion beam with an ion fluence of about 10^{15} to 10^{16} ions/cm^2. The sample temperature was kept at room temperature. The beam power was rather low (<0.3 W) and thus the beam heating of the sample was negligible in our case. RBS experiments have been performed for each sample after each annealing and ion irradiation, while the RBS-C experiments and XRR measurements were carried out for some chosen samples.

3. Results and discussions

The sample characterizations by LEED and CEMS measurements have indicated the good quality of the films (for details see [6,7]). It is summarized as follows: (1) the in situ LEED patterns of the deposited Fe_3O_4/MgO films have displayed clearly a $(\sqrt{2} \times \sqrt{2})R45°$ reconstruction typically observed for magnetite (001) surfaces, (2) the CEMS data analysis of the as-grown film suggested that the interface region is the spinel phase $Fe_{3-x}Mg_xO_4$ with $x = 0.10–0.15$ and a thickness of 2 nm, (3) the CEMS spectra have revealed that after one month's exposure to clean air an oxidized phase γ-Fe_2O_3 with a thickness of 1.5 nm was formed on the surface of magnetite films and (4) no significant changes in the

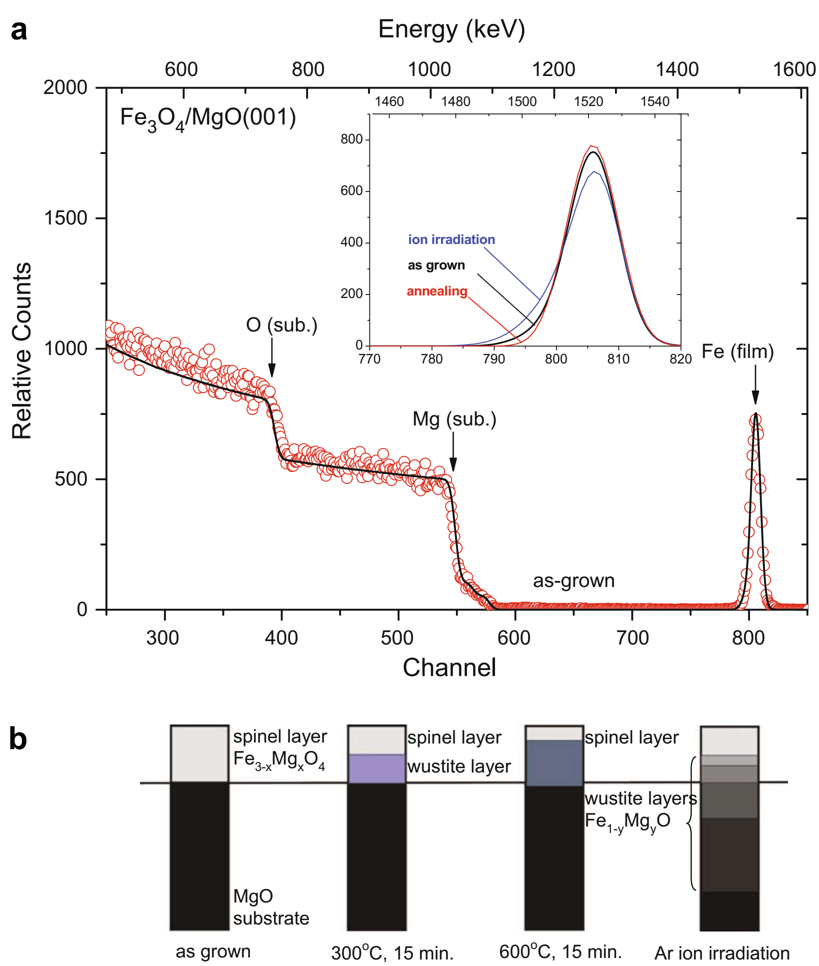

Fig. 1. (a) Random RBS (markers) and SIMNRA (lines) simulated spectra for as-grown 10 nm-thick Fe_3O_4/MgO(001) film. Inset: comparison the Fe-peak in the as-grown state with that after annealing at 600° C and after 1 MeV Ar^+ ion irradiation. RBS experiments were performed with 2 MeV He^+ ion beam and the backscattering angle of 171°. (b) Diagram of the Fe_3O_4/MgO(001)film. The solid line indicates the original separation between the film and substrate. The layer thickness of different layers is drawn proportionally with respect to the values given in Table 1. Different colors indicate different Mg and Fe composition in the layers. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)