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Report:

Metallic layers are currently a topic of extensive studies. For their applications in magnetic storage technologies or soft X-ray multilayer mirrors the understanding of the near-surface diffusion which is responsible for their stability at elevated temperatures is of great importance. This experiment was performed to study the self-diffusion in a pure iron layer and in Fe_3O_4.

Investigations of atomic mobility in structures with the truncated translational symmetry is a new field. Information on diffusion of impurity atoms on the surface has been obtained so far from macroscopic diffusion experiments or from atom-resolved scanning tunneling microscopy [1]. However, macroscopic (e.g. tracer) methods can not provide any detailed information with atomic resolution and STM is sensitive only to the upper-layer atoms different from the matrix.

The access to X-rays produced by third generation synchrotron radiation sources enable studies of dynamics in metallic systems with a new technique. The effect of total reflection of X-rays and neutrons have become established techniques of studying the structure of thin films and multilayers. The special goal of these methods is the ability to perform a depth profile analysis [2]. The combination of total reflection of X-rays with Mössbauer spectroscopy of synchrotron radiation enables the depth-selective investigation of hyperfine parameters of iron atoms [3]. The extension of the grazing incidence Mössbauer spectroscopy (GIMS) method for studying diffusion is straightforward. The unique goal of GIMS is its sensitivity just to motion of the near-surface atoms and *not to the atoms on the metallic surface*. The thickness sensitivity can be varied between about two and more than ten *nm*. This is due to the effect of total reflection, which occurs since the index of refraction for X-rays is less than one, as a strong exponential damping of the waves within the crystal in the direction normal to the surface.

Two samples have been investigated: (i) 300 *nm* thick iron (95% ^{57}Fe enriched) grown by molecular beam epitaxy on a cleaved MgO (001) substrate; the single crystal iron forms a bcc Fe epitaxial film with the (100) axis twisted 45° relative to the (100) substrate axis; (ii) 30 *nm* Fe_3O_4 grown also on a MgO (001) substrate. Preparation conditions and sample characterisation techniques are published in Ref. [4]. Samples were measured in a vacuum furnace mounted on the goniometer head.

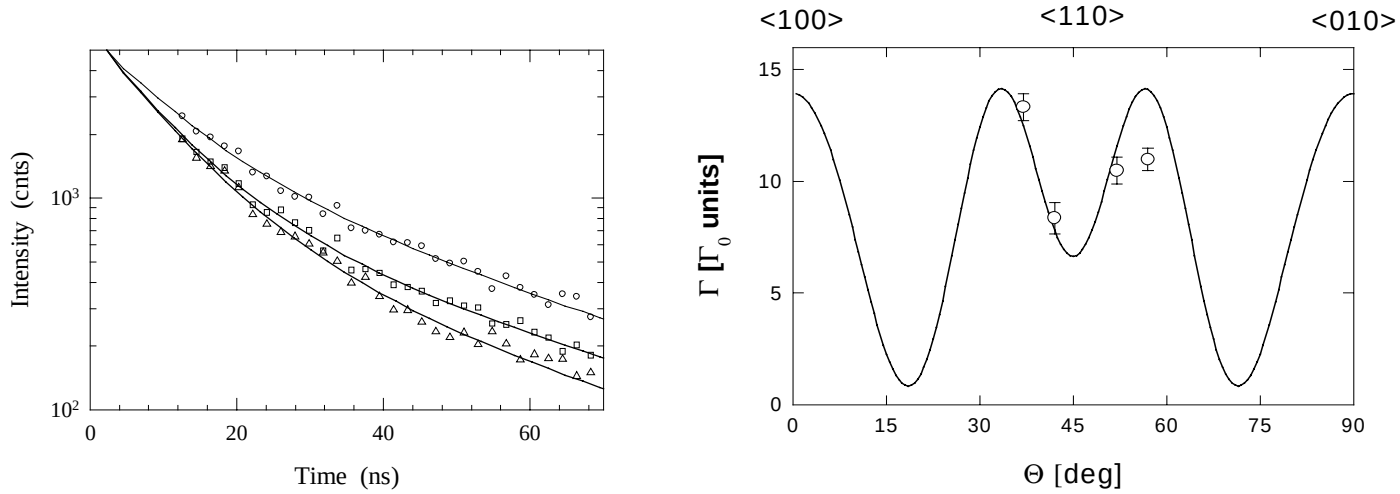

Fig. 1. (left) Intensity decay of the nuclear resonant scattered quanta from iron measured at different temperatures and at the same grazing angle of 1.66 mrad. Temperatures 1090K (circles), 1200K (squares) and 1230K (triangles); (right) intensity decay acceleration Γ at 1200K from the near-surface iron layer as a function of the azimuth angle between the SR direction and the (100) iron crystal axis.

The temperature dependence of the time decay, see Fig. 1 left, was followed yielding the accelerated intensity decay. This accelerated decay is caused by iron atoms diffusing in the near surface region of the sample [5]. Experimental data were evaluated by the EFFINO program [6]. The simplest model which could fit the experimental data consists of 2 layers and the MgO substrate. In the uppermost iron layer of about 2 *nm* thickness a noticeable decay acceleration was fitted as shown in Fig. 1 right. The second layer is formed by bulk-like iron. The diffusion coefficient of iron in the near-surface layer is one order of magnitude higher than in bulk bcc iron at the same temperature.

The intensity decay in single crystals depends on the relative orientation of the jump vector and the outgoing wave vector. This is the reason for an angular dependence of the decay constant Γ shown in Fig. 1 (right) in natural decay units in ^{57}Fe, Γ_0=1/141*ns*. A similar angular dependence was found at 1230K. This proofs that the measured effect has a *dynamical origin*. The solid line in Fig 1. is simulated for a b.c.c diffusion model with nearest-neighbours jumps between lattice sites. In addition nuclear and electronic reflectivities of the iron sample were measured (Fig.2).

The measurements on magnetite were not successful due to deterioration of the structure by interdiffusion with the MgO substrate. It has to be performed under in situ control of structure and composition.

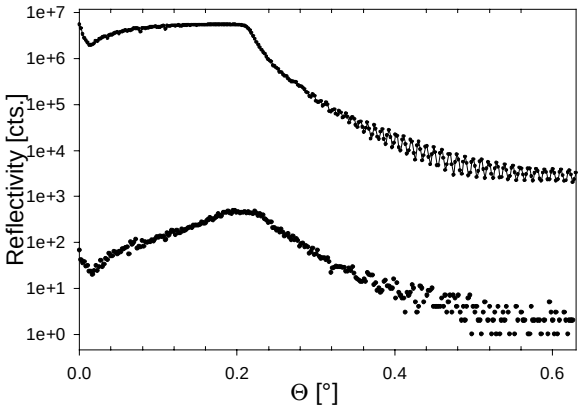

Fig. 2. From the period of Kiessig beats [3] of the electronic reflectivity the thickness of the iron layer can be estimated (upper curve). The nuclear reflectivity shows its maximum at the critical angle of the total reflection at 3.8 *mrad*. The iron sample was measured at a smaller angle (1.66 *mrad* = 0.095°) where the penetration depth in iron is about 3 *nm*.

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