Magnetic Friedel oscillation at Fe(001) surfaces: Direct observation by atomic-layer-resolved synchrotron radiation ^{57}Fe Mössbauer spectroscopy

A study of the surface and interface magnetism of 3*d* transition metals is of interest because of the essential role that magnetism plays in determining magnetic interactions and spin-transport properties of nanomagnets and magnetic heterojunctions. Over the past few decades, various techniques have advanced the studies on surface and interface magnetism. However, few experimental studies on the depth-dependent local magnetic structures of surfaces and interfaces at the atomic layer level have been carried out. This situation is a result of the difficulties encountered when performing depth-resolved studies at the uppermost surface of a metal, e.g., by scanning tunneling microscopy, or the signal arising from a relatively broad range of depth, e.g., in X-ray magnetic circular dichroism spectroscopy.

The surface magnetism of Fe(001) is a fascinating research subject for atomic-layer-resolution magnetic analysis. Theoretical studies predict 30% enhancement of the magnetic moment M_{Fe} at the surface and an oscillatory behavior with increasing depth in the individual layers, i.e., a magnetic Friedel oscillation [1]. As a relevant phenomenon, Ohnishi *et al.* theoretically predicted that hyperfine magnetic field H_{int} is reduced by 30% relative to the bulk value despite a significant increase in the surface M_{Fe} [2].

Recently, we have determined the layer-by-layer H_{int} of the Fe(001) surface by the *in situ* ^{57}Fe probe layer method with a high-brilliance synchrotron Mössbauer source [3]. In this method, a resonant isotope probe layer is embedded in a thin film prepared with a nonresonant isotope. The observed H_{int} at the nucleus provides details on the local surface magnetism.

An *in situ* measurement system was developed to observe the magnetic Friedel oscillations on the surface of Fe. It consists of a molecular beam epitaxy (MBE) chamber and an ultrahigh-vacuum measurement chamber (Fig. 1(a)). The latter chamber is equipped with a liquid helium flow cryostat and electromagnetic coils, which enables low-temperature measurements and polarized Mössbauer study using a magnetized thin film.

Fe(001) films were fabricated by alternately evaporating ^{56}Fe and ^{57}Fe from 99.94% iron-56 and 95.93% ^{57}Fe isotopic sources onto precleaned 10×10×0.5 mm^3 MgO(001) substrates under a vacuum pressure of approximately 10^{-8} Pa. A 0.8-ML-thick ^{57}Fe probe layer, t = 0.1 nm, was embedded to the depth of the N^{th} atomic layer, N = 1 to 4 and 7, below the surface. These samples are hereafter referred to as "N^{th} probe layer samples".

The experiments were performed at SPring-8 **BL11XU** using linearly π-polarized 14.4 keV Mössbauer γ-rays with a 15.4 neV bandwidth produced by a synchrotron Mössbauer source. The γ-ray beam was vertically focused by an elliptical mirror. The beam size was 15 μm(V)×1.6 mm(H) and the beam flux was about 2.9×10^4 photons/s. This beam was introduced into the measurement chamber to perform grazing incidence measurements (Fig. 1(b)).

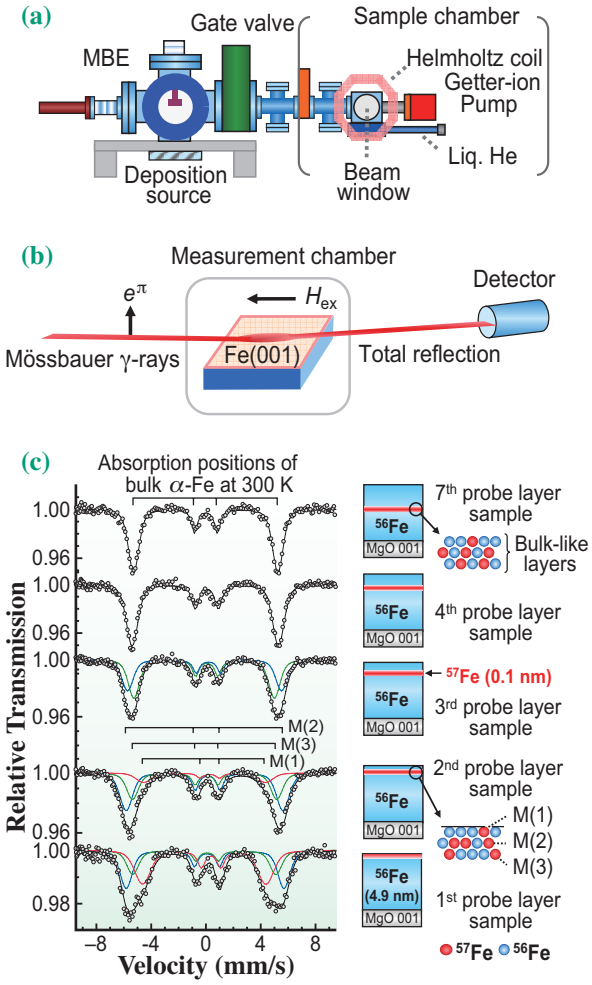

Fig. 1. **(a)** Experimental setup. H_{ex}: Magnetic field (300Oe). **(b)** Mössbauer spectra of the N^{th} probe layer samples measured at 300K. Black solid lines represent fitted curves. Red, blue, and green lines represent three different magnetic components. M(*i*) represents the magnetic component assigned to the ^{57}Fe atoms located in the i^{th} layer below the surface.

An external field of 300 Oe was applied antiparallel to the beam direction to magnetize the Fe(001) film. In this arrangement, the π-polarized incident beam interacted with the four nuclear transitions of $\Delta m = \pm 1$. The Mössbauer absorption spectra were measured by collecting the totally reflected γ-rays from the sample surface at an incident angle of 0.1° with a reflectivity of about 80%. Each spectrum was obtained within a few hours of sample preparation. Such short-time measurements significantly reduced the residual gas absorption and oxidation on the Fe(001) surfaces.

Figure 1(c) shows the Mössbauer spectra of the N^{th} probe layer samples, N = 1 to 4 and 7, recorded at 300 K. All samples showed magnetically split Mössbauer patterns. The spectra of the first, second, and third probe layer samples exhibited complex profiles composed of different magnetic components [i.e., small H_{int} (red lines, around 28 T), large H_{int} (blue lines, around 36 T), and bulk-like H_{int} (green lines, around 32 T)].

The ideal probe layer in the sample was surrounded by finely distributed ^{57}Fe atoms, which stemmed from the random deposition and surface diffusion of iron atoms during the growth process. Figure 1(c) (right) shows a conceptual diagram. In this case, if the first, second, and third layers of the iron surface have different H_{int} values, the spectra should exhibit a complex profile with multiple components. On the basis of the systematic behavior of the three components, the small H_{int}, large H_{int}, and bulk-like H_{int} represented the intrinsic hyperfine fields for the first, second, and third layers from the surface, respectively. In contrast, the spectra of the fourth and seventh probe layer samples exhibited a single magnetic component with four absorption lines, even in the presence of finely distributed ^{57}Fe atoms. This is because the hyperfine fields of the neighboring layers in these depth regions are bulk-like, and the overlapping subspectra result in a simple absorption profile. The prominent subspectrum with the largest percent area in the N^{th} probe layer sample was assigned to the spectrum characterizing the ^{57}Fe atoms located in the N^{th} atomic layer from the surface.

The experimentally determined layer-by-layer H_{int} exhibited a marked decrease at the surface and an oscillatory decay toward the bulk value. This behavior was successfully reproduced by theoretical calculations (Fig. 2). The result provides the first experimental evidence for magnetic Friedel oscillations, which penetrate several layers from the Fe(001) surface. Theoretically, the oscillatory decay of H_{int} should be strongly coupled with the Friedel oscillation of M_{Fe}, which is caused by the surface electronic structure with a large spin imbalance and *d*-band narrowing [1-3]. A schematic diagram of the magnetic Friedel oscillations in M_{Fe} and H_{int} is shown in Fig. 3.

This study yielded a clear answer to the mystery of the surface magnetism of iron, which has been discussed since the 1980s. In the future, the *in situ* ^{57}Fe probe layer method with a synchrotron Mössbauer source should facilitate additional studies on the surface and interface magnetism in advanced magnetic and spintronic materials and devices.

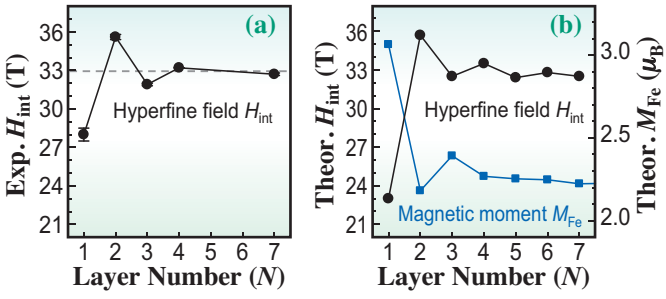

Fig. 2. Plots of the experimental and theoretical layer-by-layer H_{int} and M_{Fe}.

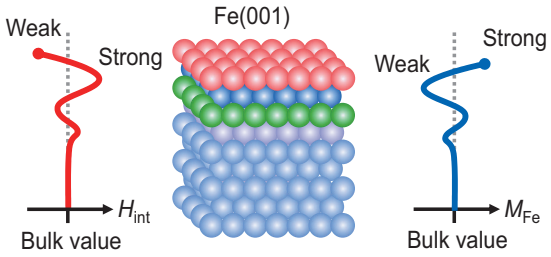

Fig. 3. Schematic diagram of the observed magnetic Friedel oscillation of Fe(001) surface.

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