Magnetic Friedel Oscillation at Fe(001) Surfaces: Direct Observation by Atomic-Layer-Resolved Synchrotron Radiation ^{57}Fe Mössbauer Spectroscopy

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The study of the surface and interface magnetism of 3^{d} transition metals is of interest due to the essential role that magnetism plays in determining magnetic interactions and spin-transport properties of nanomagnets and magnetic hetero junctions. Over the past few decades, various techniques have advanced the research on surface and interface magnetism. However, few experimental studies have investigated the depth-dependent local magnetic structures of surfaces and interfaces at the atomic layer level. This is due to the difficulties encountered when performing depth-resolved studies at the uppermost surface of a metal, such as with scanning tunneling microscopy, or due to the signal arising from a relatively broad depth range, e.g., with x-ray magnetic circular dichroism spectroscopy.

The surface magnetism of Fe(001) is a fascinating research subject for atomic-layer-resolved magnetic analysis. Theoretical studies predict a 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 related phenomenon, Ohnishi et al. theoretically predicted that the 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 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 non-resonant isotope. The observed H_{int} at the nucleus provides details on the local surface magnetism.

Fe(001) films were fabricated by alternatively evaporating ^{56}Fe and ^{57}Fe from 99.94% iron-56 and 95.93% ^{57}Fe isotopic sources onto pre-cleaned 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 Nth atomic layer below the surface where N = 1 to 4 and 7. These samples are hereafter referred to as "Nth probe layer samples".

The experiments were performed at the BL11XU beamline of SPring-8 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 approximately 2.9×10^4 photons/s. This beam was introduced into the measurement chamber to perform grazing incidence measurements (Fig. 1a). 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 approximately 80%. Each spectrum was obtained within a few hours of sample preparation. Such rapid measurements significantly reduced the residual gas absorption and oxidation on the Fe(001) surfaces.

Figure 1b presents the Mössbauer spectra of the Nth 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).

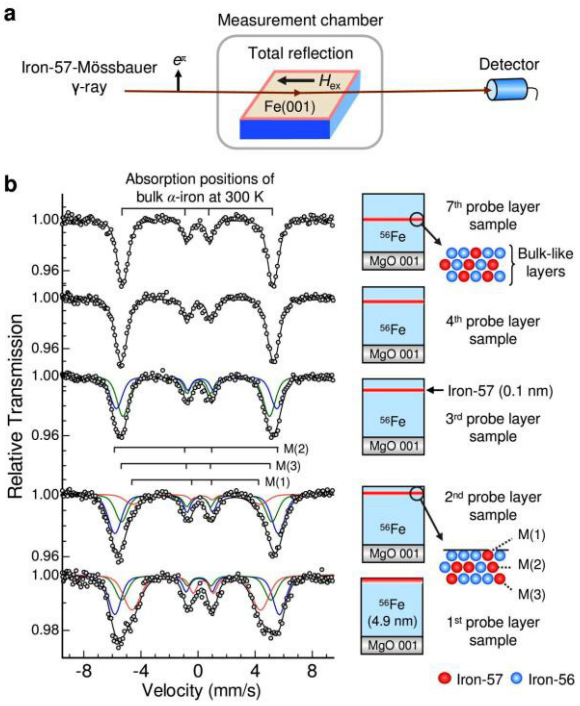

Figure 1. (a) Experimental setup. (b) Mössbauer spectra of the Nth probe layer samples measured at 300 K. Black solid lines represent the fitted curves. Red, blue, and green lines represent the three different magnetic components. M(i) represents the magnetic component assigned to the ^{57}Fe atoms located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

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 1b (right) shows a conceptual diagram of the process. In this case, if the first, second, and third layers of the iron surface have a different H_{int} the spectra should exhibit a complex profile with multiple components. Based on the systematic behavior of the three components, the small H_{int}, large

H_{int}, and bulk-like H_{int} represent 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 at these depths 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. Such a behavior was successfully reproduced by theoretical calculations (Fig. 2). This result provides the first experimental evidence for the magnetic Friedel oscillations, which penetrate several layers below 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, 2, 3].

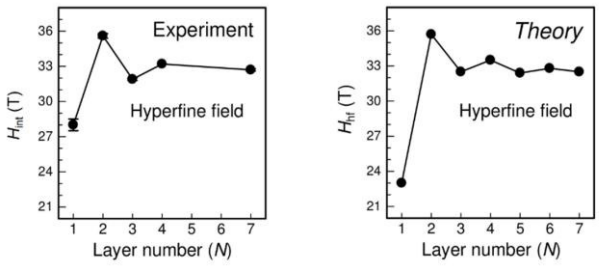

Figure 2. The experimental and theoretical layer-by-layer hyperfine magnetic fields.

In summary, the surface magnetism of Fe(001) was studied in an atomic layer-by-layer manner by using the in situ ^{57}Fe probe layer method with a synchrotron Mössbauer source. The observed H_{int} exhibited a marked decrease at the surface and an oscillatory behavior with increasing depth in the individual upper four layers below the surface. 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.

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References

1. C. S. Wang and A. J. Freeman, PRB **24** (1981)4364.
2. S. Ohnishi et al: PRB **28** (1983) 6741.
3. T. Mitsui et al: PRL 125 (2020) 236806.