Thickness dependent crystallographic transition in Fe/Ni multilayers

Lennart Häggström¹, Inna Soroka² and Saeed Kamali³
¹Department of Physics and Materials Science and ²Department of Materials Chemistry, Uppsala University, Box 530, 751 21 Uppsala, Sweden
³Department of Applied Science, University of California, Davis, Cal. 95616, USA
E-mail: lennart.haggstrom@fysik.uu.se

Abstract The crystallographic transition between bcc- and fcc-structures has been studied in Fe/Ni multilayers with Mössbauer spectroscopy and X-ray diffraction. Layered structures have been confirmed and the structures depends both on individual layer thickness and the ratio of the constituents. Two sets of samples are made: the Fe layer thickness kept constant to 12 monolayer (ML) and 24ML, while the Ni layer thicknesses varied from 3 to 12ML. When the Ni layer is thin, the Fe/Ni multilayers have the bcc phase. When the thickness of Ni layer reaches 6ML and larger, a gradual transition to the fcc phase occurs. Mössbauer spectroscopy studies confirm the existence of a magnetic Fe bcc phase with in-plane magnetization in all samples, two magnetic fcc phases with fields of 28 T and 10 T and small amounts of non-magnetic fcc phases in the thicker films. The observed behaviour can be attributed to the existence of two types of interfaces which are not necessary overlapping each other: An elemental interface between Fe and Ni and a structural interface between fcc and bcc phases.

1. Introduction
The 3d transition metals have attracted large attention and FeNi alloys are of particular interest due to the so called Invar effect [1] coupled to a phase transformation. While bcc Fe is ferromagnetic at room temperature (RT), fcc Fe can have different magnetic states such as ferromagnetism, antiferromagnetism, and can even be non-magnetic. Fcc iron is unstable at RT and can be stabilized in the form of thin layers or nanoparticles on fcc substrates or matrices. There are a number of studies where Fe is stabilized in a fcc phase in a Cu matrix [2,3]. Layered system of Fe on Ni single crystal has also been studied [4]. Mössbauer spectroscopy (MS), is a powerful technique for studying these systems because the magnetic hyperfine fields are different for Fe atoms in bcc and fcc environments. In the past, MS has been used to investigate Fe/Ni layered systems [5]. In almost all the previous studies of this system, a thick Ni layer has been used, as either substrate or buffer layer, and then a thin Fe layer has been grown onto this Ni layer to establish a fcc structure of Fe. The present work is dedicated to the investigation of the crystallographic transition from bcc to fcc phase in Fe/Ni multilayers. We stabilize first the bcc phase with thick Fe and thin Ni layers, and then the fcc phase is obtained by increasing the Ni layer thickness while the Fe layer thickness is kept constant.

2. Experimental
Samples with x ML of Fe and y ML of Ni layers were grown (samples denoted hereafter by x/y ) in a UHV chamber on single crystal MgO(001) substrates by dc magnetron sputtering. Separate Fe (99.95 % purity) and Ni (99.99 %) targets are used. The base pressure was lower than 2 · 10⁻⁹ Torr and the operating pressure of Ar gas was 2.4 mTorr. The deposition rates were 0.30 Å/s and 0.59 Å/s for Fe
and Ni, respectively. The optimal substrate temperature was determined to be 230°C, as deduced from structural analysis. There was no buffer layer and the growth always started with Fe layers and on top a cap with 10 Å thick Pd layer to prevent oxidation was deposited. Two sets of samples were grown: in one set the thickness of Fe layer is kept constant at 12 ML while the thickness of Ni layer varied from 3 to 12 ML. The second set consists of Fe/Ni layers with constant thickness of Fe layer, 24 ML, while the Ni layer thickness is changed from 5 to 12 ML. The number of repetitions of 1 bilayers is 50 and 25 for the first and the second set of samples, respectively. The structure, bilayers thicknesses as well as total thicknesses of all films were determined by X-ray diffraction (XRD) and X-ray reflectivity (XRR) measurements using a Siemens D5000 diffractometer with CuKα radiation. 57Fe Conversion Electron Mössbauer Spectroscopy (CEMS) was performed at room temperature with 57Co in a Rh matrix as the source. A gas flow (He & CH4) detector was used. In all measurements, the direction of the incoming photons was perpendicular to the sample plane. All isomer shifts are given with respect to α-iron at room temperature.

3. Result and discussion

3.1 XRR and XRD measurements

As seen in the XRR patterns, Figure 1(a and c), superlattice peaks appear, indicating that the films have layered structure in all samples. Also, the finite size oscillations (Kiessig fringes) are clearly seen except for the thickest Ni layers. This confirm the good quality of the surfaces and the interfaces between the substrates and the films. With high angle XRD measurements the structural changes as function of Ni layer thickness are obtained. The XRD spectra for the multilayers with thin Ni layers (12/3) and 12/3.5) have only one diffraction peak (Figure 1b), which corresponds to the (002) plane in bcc Fe.

![Figure 1](image-url)

The crystal coherence length (ζ), obtained from Full Width Half Maximum (FWHM) of the Fe bcc peak, is found to be about 110 Å for both samples. Thus, those films can be considered as monocrystals. The structure started to change dramatically when the Ni layer thickness become larger than 6ML. Additional diffraction peaks from (022) and (002) fcc Ni phase appears in the XRD spectra for 12/6 and 12/12. Moreover, the FWHM of the (002) bcc peaks strongly increased and their intensities decrease. This indicates that the films became polycrystalline and consist of bcc and fcc phases, while the layered structure is still kept. In the second set of samples, where the Fe layers are twice as thick as compared to the films in the first set, the structural changes have different behaviour. As seen in the Figure 1d, the 24/5 film has one XRD peak that corresponds to the (002) plane of the Fe bcc phase with ζ=100Å. When the thickness of the Ni layer is increased to 6 ML, the (002) bcc peak became broader (ζ~75Å) and its intensity decreased. At the same time additional small peaks at higher angles appear. Those peaks become more pronounced in the XRD spectra of the 24/7 film, where the splitting of the (002) peak is observed. The origin of those peaks is not clear, however we can assume that the film consists of several regions where the lattice parameters...
and crystalline orientation differ from each other. Note that the 24/7 film has well defined layered structure, according to the XRR measurements. Finally, in the XRD pattern for the sample 24/12 two wide peaks which correspond to (002) bcc Fe and (022) fcc Ni are present. Accordingly, in the film where the Ni amount is rather high, two phases most probably textured in the directions [001] bcc and [011] fcc are formed. Thus, the structure of the Fe/Ni multilayers change from monocrystalline bcc phase to polycrystalline bcc+fcc phases by increasing the thickness of Ni layer keeping the Fe layer thickness unchanged. From the XRR measurements, the average film monolayer thickness as a function of Fe/Ni ratio can be obtained. For the first set of samples this thickness increases with the Ni content. This result is in good agreement with the increased amount of the fcc phase. However, for the second set, the average film monolayer thickness does not depend much on the composition and remains close to bulk Fe value, 1.43Å.

3.2 Mössbauer spectroscopy

The Mössbauer spectra and fitting results are displayed in Figure 2. As can be seen, the hyperfine fields $B_{hf}$ can be grouped into three well separated categories, one high field value of 33.4 T (distribution width $\sigma \approx 0.9$ T), one intermediate field value of 28.2 T ($\sigma \approx 1.2$ T) and one low field value of 10.9 T ($\sigma \approx 2.0$ T) for all samples.

Beside these spectral patterns there is also a broadened singlet line in some of the spectra. It has recently been shown that the Fe/Co superlattices can be analyzed using a local alloy model [6]. We adopt the same procedure of analysis in the present case of Fe/Ni multilayers. Johnson et al. [7] have published Mössbauer result at RT of the Fe-Ni alloys (Figure 3). There is a transformation between the bcc and the fcc structures for these alloys at a Ni concentration between 24% and 34%. In the bcc structure, for lower Ni concentration, the hyperfine field is between 33.0 T and 34.3 T. At the crystal transformation a two phase region exists where the hyperfine fields for Fe in the fcc-alloys vary between 0 T and about 25 T. For higher concentration, in the fcc phase, the hyperfine field varies between 28.2 T and 30.8 T. A hyperfine field gap (30.8 T-33.0 T) thus exists between the bcc- and fcc-alloys. It is, therefore, tempting to ascribe the high field component of 33.4 T to Fe in the bcc structure with a Ni concentration less than 30% in the surrounding of each Fe atom, the intermediate field of 28.2 T, to Fe in the fcc structure with a Ni concentration above 40% and the low field of
10.9 T to Fe in fcc-type of structure with a Ni concentration of \( \approx 30-40\% \). The broadened singlet line with zero field would then be assigned to the fcc structure with a narrow Ni concentration around 30\%. This description is further supported by the findings of a hyperfine field of 28.5 T for Fe in a 5 ML thick fcc-Fe(001) film on a fcc Cu(001) substrate [3] and fields of about 28 T and 8 T for Fe fcc films with the thickness of about 10 Å in Ni/Fe/Ni trilayers [8].

![Figure 3](image3.png)  
**Figure 3.** The \(^{57}\)Fe hyperfine field at room temperature in the Fe-Ni alloys [7]. The dashed vertical line marks the crystallographic transformation between bcc (left part) and the fcc (right part) structures. Note the hyperfine gap (30.8 T - 33.0 T) between the fields in bcc- and fcc-alloys, respectively.

Figure 4 displays the spectral intensity of the bcc and the total fcc components as a function of Fe/Ni ratio. For a ratio above 3.5 the multilayers have a dominant bcc structure while when the ratio goes towards 1 the fcc structure dominates. The observed behavior can be attributed to the existence of pure bcc and fcc phases and structural interfaces in between. These interfaces do not need, however, to overlap with the elemental interfaces between Fe and Ni. In the boundary between bcc-fcc phases the isomer shift for the fcc pattern shows a shift with respect to the bcc pattern of -0.12 mm/s for the FeNi-alloys [8]. The bcc-fcc shift for the Fe/Ni multilayers is smaller, about -0.06(2) mm/s. In the multilayers with the larger Ni layer thickness the structure gradually changes between the two phases and there can appear local Fe surroundings which have rather distorted bcc and fcc structures. This may explain why the isomer shift values for Fe in the superlattices do not exactly match the alloy values. The line intensity ratios for the magnetic six-line pattern are found very close to 3:4:1:1:4:3. Since the \( \gamma \)-ray direction was perpendicular to the sample plane, these intensity ratios indicate that the average magnetic moment direction is oriented within the sample plane. This is expected from the shape anisotropy of the multilayer films.

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