

Supplementary section: Nanometric Self-diffusion of Fe: effect of grain size

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Separating interface roughness, Fe-Pt intermixing from interdiffusion in bilayers:

We present simulation plots with increasing surface roughness for the Fe₂ and Fe₅ samples without considering any interdiffusion. The plots clearly indicate that the changes in the reflectivity profile are quite different as compared to that due to interdiffusion at the interfaces. The effect of increasing Fe-Pt interface roughness with respect to the NR data (solid symbols) for Fe₅ sample at t_0 and t_5 and Fe₂ at t_{90} are shown in Fig. 1(a and b).

The purpose in Fig. 1(a) is to replicate the differences in the t_5 data by considering a change only in the surface or interface roughness (the effect of these two roughness factors surface or interface roughness is very similar on the profile) from the case that is considered in the fit to the t_0 data. The roughness is increased from 1.0 to 3.0 nm. We have not considered any interdiffused layer ($t_d=0.0$ nm) in simulating the reflectivity profiles and have kept all other parameters unchanged. Note that the Fe-Pt intermixing factor is not sensitive enough to be considered in these bilayers and was kept constant at 0.5 nm. One can easily see that it is not possible to get any reasonable fit to the t_5 (or to the t_{90} data) data, simply by considering an increased surface or Fe-Pt interface roughness. The discrepancy is distinct around $Q_z=0.35$ nm⁻¹ to $Q_z=0.55$ nm⁻¹ for the Fe₅ sample. This difference obviously is more pronounced in the cases of further annealing times.

For the Fe₂ sample in Fig. 1(b), one can see a shift in the oscillation period with annealing. The second peak is shifted from 0.5 nm⁻¹ (black curve) to 0.55 nm⁻¹ (magenta curve). This change in periodicity cannot be replicated by considering an increased roughness but only by considering an interdiffusion in Fe *i.e.*, a change in the SLDs of the layer constituents. A reasonable fit is possible only and only when a Fe interdiffused layer is considered. Thus we have demonstrated that the changes in the NR patterns after annealing, may be small but the effect on the patterns due to Fe-Pt interface roughness is significantly different from the changes due to Fe interdiffusion. Please note that the interface roughness from the X ray fits clearly show that there is hardly any change due to annealing. Since with X rays we are sensitive only to the Fe-Pt roughness and not to the Fe interdiffusion, these values can roughly be used as the starting parameters in the fitting of the NR profiles and they do not change.

Thus given the fact that the interface roughness remains unaffected upon annealing and the Fe-Pt intermixing being negligible in the bilayer samples; the only contributing factor

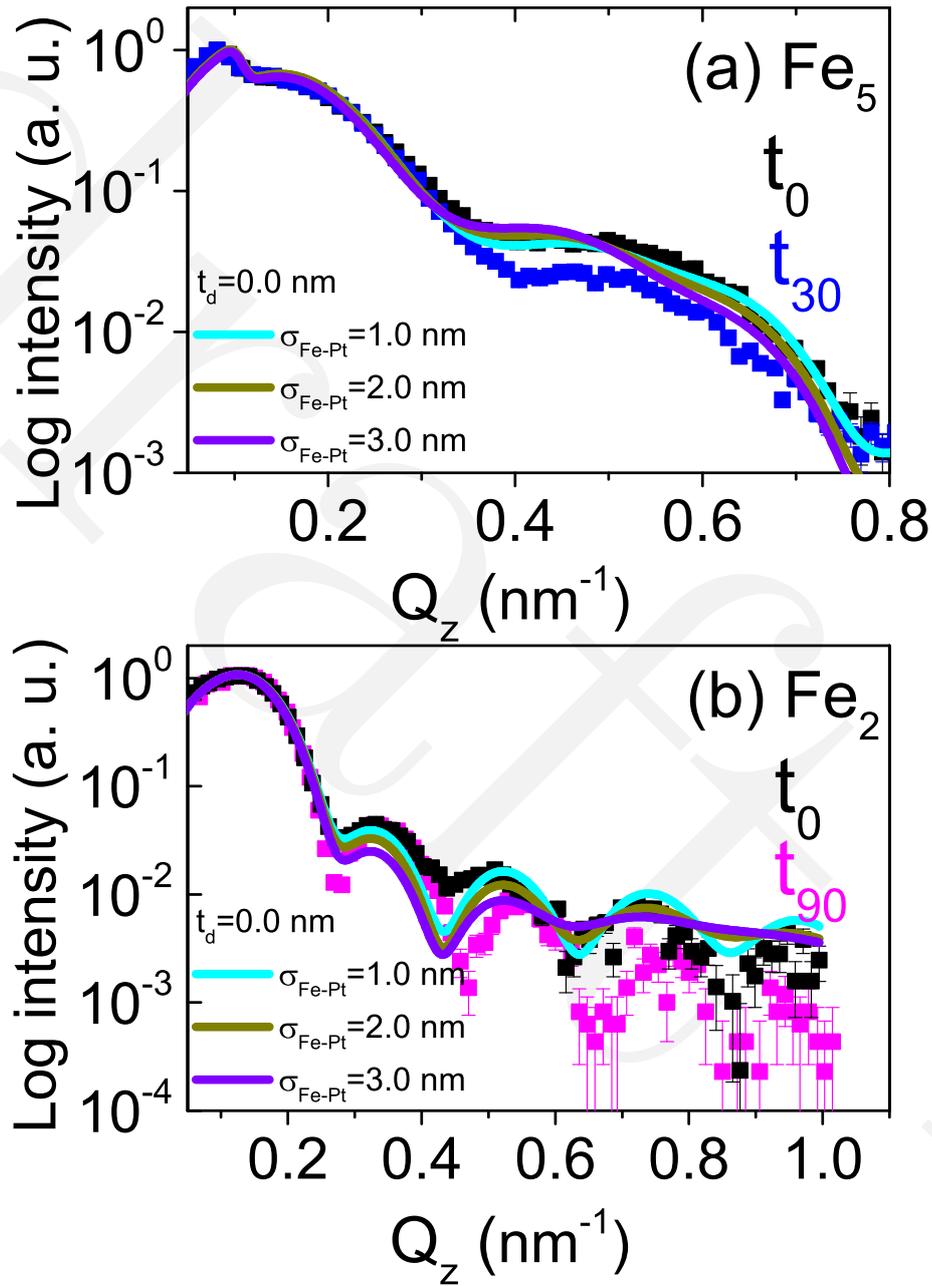


FIG. 1: (Color online) NR simulated patterns versus Q_z corresponding to the (a) Fe₅ bilayer and (b) Fe₂ bilayer for various surface roughness σ while the interdiffused layer thickness $t_d=0$. They show a change in shape and decrease of the intensities which is different than considering finite t_d values.

to be considered is the Fe interdiffusion. Overall, the two factors, (i) the Fe interdiffusion and (ii) Fe-Pt intermixing affects the reflectivity profile differently. Identification of these two is possible due to large nuclear SLD contrasts between (a) ^{56}Fe ($7.8 \times 10^{-6} \text{ \AA}^{-2}$) and ^{57}Fe ($2.7 \times 10^{-6} \text{ \AA}^{-2}$) and that between (b) the Fe-Pt intermixed layer ($3.3 \times 10^{-6} \text{ \AA}^{-2}$) and Pt ($6.8 \times 10^{-6} \text{ \AA}^{-2}$) layer, respectively.

Separating interface roughness, Fe-Pt intermixing from interdiffusion in multilayer:

To prove the point of interface roughness, intermixing and interdiffusion in the multilayer we show in Fig. 2(a) the simulated reflectivity profiles with increased roughness from 1.0 nm to 2.0 nm corresponding the un-annealed (t_0) sample and with an increased interdiffused layer thickness t_d from 0.0 nm to 2.9 nm corresponding to the maximum annealed (t_{3000}) sample. Firstly, one should note the decrease in the intensity of the 2^{nd} order Bragg peak with an increased roughness (black and gray curves) and secondly the broadened shape of the 1^{st} order Bragg peak (orange and pink curves) which are affected by an increased interdiffusion. Thus, for such a combination of layer thickness, the increase in interface roughness reduces the 1^{st} order Bragg peak to some extent and the 2^{nd} order Bragg peak drastically, while the increase in interdiffused layer affects the intensity and shape of both Bragg peaks. The changes in the top Pt layer SLDs with annealing can also be separately accounted for, separate from the rest of the isotopic periodic stack.

It can be seen in Fig. 2(b) that Fe-Pt intermixing, which occurs mainly after the first annealing step, has also contributed to the decrease of the Bragg peaks. Here we show the effect of Fe-Pt intermixing when $t_d=0$ nm and 2.9 nm, respectively. The 1^{st} order Bragg peak is reduced to 78% of the initial peak intensity considering only an increased Fe-Pt intermixing but without any interdiffusion (blue curve). The same peak is reduced to 42% of its initial intensity without considering any further Fe-Pt intermixing (green curve) but with a maximum interdiffusion possible. In the absence of Fe-Pt intermixing it is not possible to match the corresponding changes in intensity even if we consider sufficient Fe interdiffusion to occur. The intensity difference of the Bragg peak between the best fit curve at t_0 (black curve) and the blue curve is similar to the difference between the best fit curve at t_{3000} (orange curve) and the green curve. Thus the measured diffusion coefficients are exclusively due to grain boundary diffusion and not due to the Fe-Pt intermixing. In this

case, it is therefore more reasonable to calculate the diffusivity from the thickness of the interdiffused layer rather than from the decrease in the Bragg peak intensity. This has in fact taken care of the error factor that would have occurred in the calculation of diffusivity due to the factor of Fe-Pt intermixing.

Furthermore, we explain the procedure followed in determining the grain boundary diffusion exclusively from the intermixing of Pt with the iron films. The two factors, (i) the Fe interdiffusion and (ii) Fe-Pt intermixing affects the reflectivity profile differently. Note that here we do not consider any change in the interface roughness (which would have also affected the Bragg peak intensity) due to annealing following the results of the X ray data fits. Identification of these two is possible due to large nuclear SLD contrasts between (a) ^{56}Fe ($7.8 \times 10^{-6} \text{ \AA}^{-2}$) and ^{57}Fe ($2.0 \times 10^{-6} \text{ \AA}^{-2}$) and that between (b) the Fe-Pt intermixed layer ($3.3 \times 10^{-6} \text{ \AA}^{-2}$) and Pt ($5.6 \times 10^{-6} \text{ \AA}^{-2}$) layer, respectively.

One can separate the simulated profiles into two regions: region I and region II. Region I (below $Q_z \approx 0.5 \text{ nm}^{-1}$) largely defines the critical angle and the corresponding intensity variation. In region II (above $Q_z \approx 0.5 \text{ nm}^{-1}$) the effect of Fe interdiffusion can be seen mostly from the decrease in the Bragg peak intensities and the changing shape of the peak. The effect of Fe-Pt intermixing (t_{intermix}) and Fe interdiffusion (t_d) has been shown separately in Fig. 3(a,b). The respective t_{mixing} values on top in Fig. 3(a) are those obtained from the fits to the data (SLD profiles in the paper) but considering $t_d=0 \text{ nm}$. Following the simulated patterns, one can see a systematic decrease in the intensity in region I from t_0 to t_{3000} . The intensities in region II remain very similar for the annealed samples. This indicate changes in the top layer SLDs or Fe-Pt intermixing. Similarly, in Fig. 3(b), we show the simulations where we allow only the respective values of t_d at each annealing time (from the SLD profiles in the paper) while keeping the t_{intermix} layers on top unchanged from that of the t_0 sample. Here one can see that there is very little change in the intensity in region I and most of the changes are in region II for the annealed samples. It can therefore exclusively owed to Fe interdiffusion.

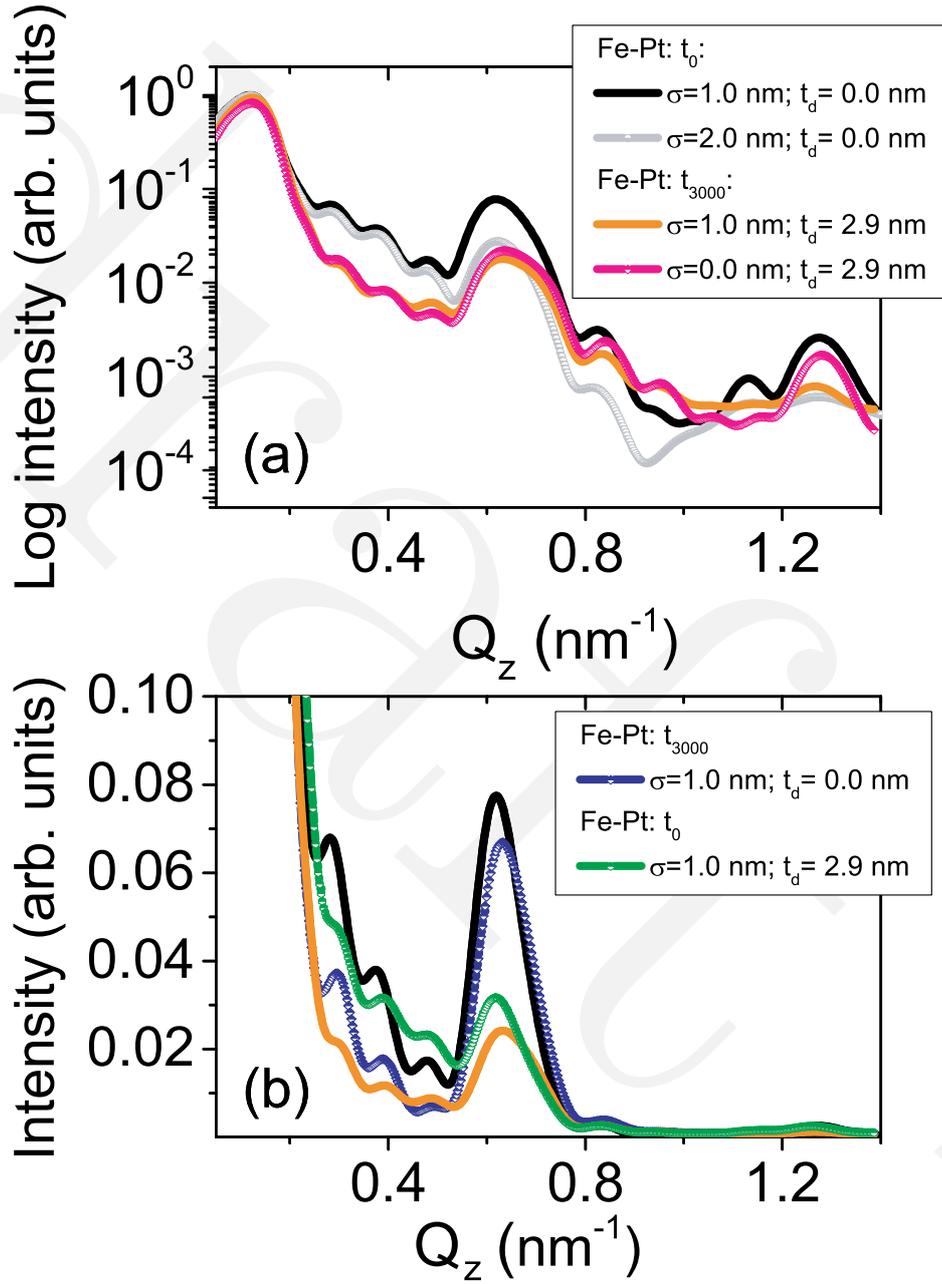


FIG. 2: (Color online) (a) NR simulated patterns versus Q_z corresponding to the $\text{Fe}_{N=5}$ multilayer for various surface roughness σ and interdiffused layer thickness t_d to show the change in shape and decrease of the two Bragg peak intensities. (b) The effect of the top Fe-Pt layer intermixing on the Bragg peak intensity with and without interdiffusion. Here, Fe-Pt: t_0 is depicting the situation of the un-annealed Fe-Pt interfaces whereas Fe-Pt: t_{3000} is the maximum annealed condition of the same.

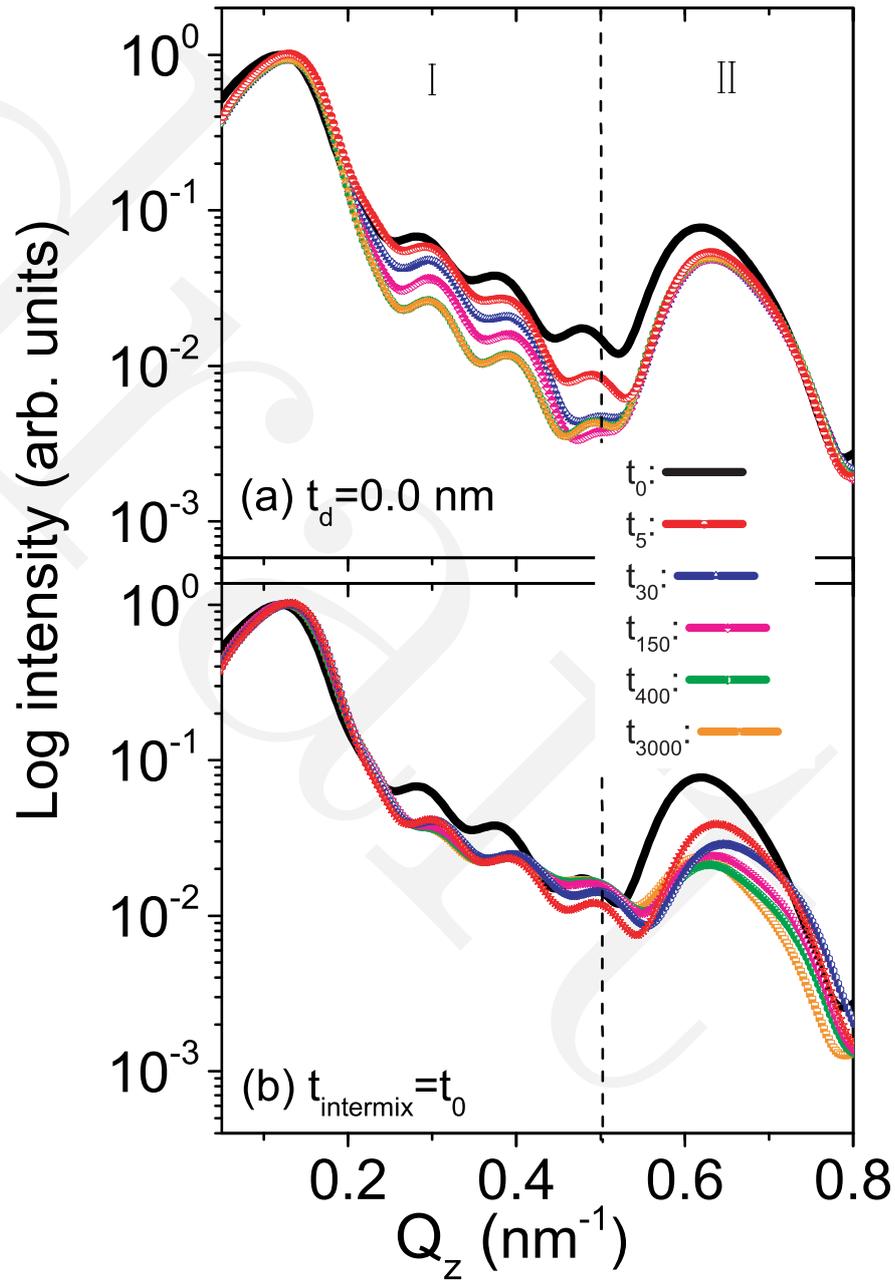


FIG. 3: (Color online) (a) NR simulated patterns versus Q_z corresponding to the $Fe_{N=5}$ multilayer for the respective Fe-Pt intermixing ($t_{intermix}$) layer at different annealing times while the interdiffused layer thickness $t_d = 0$ nm. (b) The effect of the respective Fe interdiffusion (t_d) layer at different annealing times as the $t_{intermix}$ layers are fixed to the values obtained for the t_0 sample.