Supplemental Material to "Nuclear resonant surface diffraction of synchrotron radiation"

Kai Schlage,[1] Liudmila Dzemiantsova,[1,2] Lars Bocklage,[1,2] Hans-Christian Wille,[1] Matthias Pues,[3] Guido Meier,[2,4] and Ralf Röhlsberger[1,2]

[1] *Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg*
[2] *The Hamburg Centre for Ultrafast Imaging, Luruper Chausee 149, 22671 Hamburg*
[3] *Institut für Nanostruktur- und Festkörperphysik, Jungiusstr. 11, 20355 Hamburg*
[4] *Max Planck Institute for the Structure and Dynamics of Matter, Luruper Chaussee 149, 22761 Hamburg*

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1. ALIGNMENT OF NANOSTRIPE ARRAYS VIA NON-RESONANT GISAXS PATTERNS

A perfect alignment can be recognized from a symmetric scattering pattern on the 2D x-ray area detector. An azimuthal angular misalignment of only 1° results in a drastic change, leading to an asymmetric shape of the diffraction pattern. A series of corresponding diffraction patterns and simulations is shown in the supplement (Fig. S1).

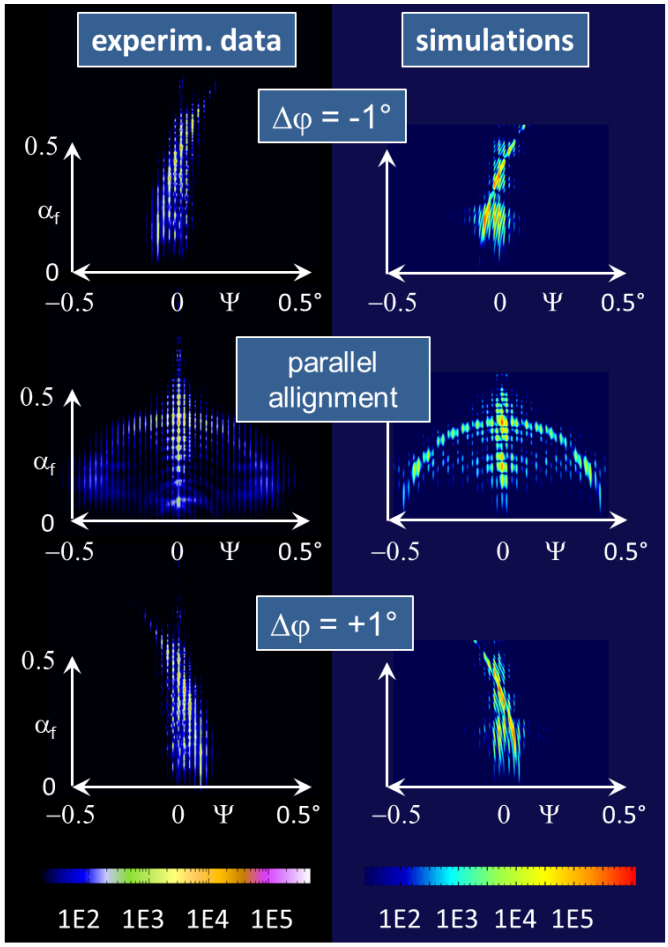

FIG. S1. 2D non-resonant diffraction data for different azimuthal orientations $\Delta\varphi$ of the wire axis relative to the beam at a constant incident angle α of 0.4°. Only a parallel allignment of the nanostripes results in a symmetric diffraction pattern. The simulation of the data sets was done with the software packages IsGISAXS [S1] and confirms the dimension of the Py stripes as intended by the lithography process. The pad structure is neglected in the simulations.

2. KINEMATICAL DESCRIPTION OF THE MEASURED TIME SPECTRA, DERIVATION OF EQ. (1)

We assume nanostripes that exhibit an antiferromagnetic order as described in the main text. The sample is illuminated under grazing angles in Faraday geometry so that $\mathbf{k_0} \parallel \mathbf{m}$, where $\mathbf{m}$ denotes the magnetization direction of the nanostripes.

In oder to describe the time spectra of nuclear resonant diffraction we start with the scattering matrix for grazing incidence reflection from a sample with an in-plane magnetization [S2, S3]:

$$\mathbf{N}(\omega) = \frac{3}{16\pi} \begin{pmatrix} F_{+1} + F_{-1} & -i(\mathbf{k}_0 \cdot \mathbf{m})\,(F_{+1} - F_{-1}) \\ i(\mathbf{k}_0 \cdot \mathbf{m})\,(F_{+1} - F_{-1}) & F_{+1} + F_{-1} \end{pmatrix} \tag{S1}$$

where $F_{+1} = F_{+1}(\omega)$ and $F_{-1} = F_{-1}(\omega)$ are the functions that describe the energy dependence of nuclear magnetic dipole oscillators for transitions with $\Delta m = +1$ and $\Delta m = -1$, respectively. Thus, for the contributions from the two sets of stripes with $\mathbf{k_0} \uparrow\uparrow \mathbf{m}$ and $\mathbf{k_0} \uparrow\downarrow \mathbf{m}$ we obtain two different scattering amplitudes resulting from the evaluation of the scalar product $\mathbf{k}_0 \cdot \mathbf{m}$ in eq. (S1) :

$$\mathbf{N}(\omega)_{\uparrow\uparrow} = \frac{3}{16\pi} \begin{pmatrix} F_P & -i\,F_M \\ i\,F_M & F_P \end{pmatrix} \quad \text{and} \quad \mathbf{N}(\omega)_{\uparrow\downarrow} = \frac{3}{16\pi} \begin{pmatrix} F_P & i\,F_M \\ -i\,F_M & F_P \end{pmatrix} \tag{S2}$$

with $F_P = F_{+1} + F_{-1}$ and $F_M = F_{+1} - F_{-1}$. If these contributions arise from laterally separated structures as in case of our sample, one has to take a spatial phase factor of $e^{i\mathbf{q}\cdot\mathbf{R}}$ into account when summing up the scattering from both parts. For simplicity we consider here scattering in the kinematical approximation only. Assuming the incident polarization given by the two-component polarization unit vector $\mathbf{A}_0$, the scattered amplitude is :

$$\mathbf{A}_S = \mathbf{N}(\omega)_{\uparrow\uparrow}\,\mathbf{A}_0 + e^{i\mathbf{q}\cdot\mathbf{R}}\,\mathbf{N}(\omega)_{\uparrow\downarrow}\,\mathbf{A}_0 \tag{S3}$$

where $\mathbf{q}$ is the momentum transfer and $\mathbf{R}$ is the spatial separation of the objects. For incident horizontal (σ) polarization $\mathbf{A}_0 = (1, 0)$ we obtain (omitting the prefactor $3/16\pi$) :

$$\mathbf{A}_S = \begin{pmatrix} F_P \\ i\,F_M \end{pmatrix} + e^{i\phi} \begin{pmatrix} F_P \\ -i\,F_M \end{pmatrix} = \begin{pmatrix} (1 + e^{i\phi})F_P \\ i\,(1 - e^{i\phi})F_M \end{pmatrix}, \tag{S4}$$

where we have set $\phi = \mathbf{q} \cdot \mathbf{R}$ for the spatial phase. Assuming unpolarized detection of the scattered radiation, the measured intensity is given by the modulus squared of $\mathbf{A}_S$:

$$I(t) = |\tilde{\mathbf{A}}_S|^2 = |\tilde{F}_P(t)|^2\,\cos^2\left(\frac{\phi}{2}\right) + |\tilde{F}_M(t)|^2 \sin^2\left(\frac{\phi}{2}\right) \tag{S5}$$

where the tilde symbol denotes the Fourier transform from the energy into the time domain. In case of the first pure nuclear diffraction peak (diffraction order 1/2), we have $\phi = \pi$, resulting in $I_{1/2} = |\tilde{F}_M|^2$ while for the specular peak (zero order diffraction) with $\phi = 0$, one obtains $I_0 = |\tilde{F}_P|^2$. Thus, the spatial phase ϕ acts as a mixing angle for the two time spectra $|\tilde{F}_P(t)|^2$ and $|\tilde{F}_M(t)|^2$. This explains qualitatively the different shape of the time spectra shown in Fig. 4 of the manuscript, as illustrated below in Fig. S2.

[S1] R. Lazzari, Appl. Cryst. **35** 406 (2002)

[S2] R. Röhlsberger, *Nuclear Condensed Matter Physics using Synchrotron Radiation*, Springer Tracts in Modern Physics, Vol. 208 (Springer, Berlin 2004).

[S3] R. Röhlsberger, J. Bansmann, V. Senz, K. L. Jonas, A. Bettac, K. H. Meiwes-Broer and O. Leupold, Phys. Rev. B 67, 245412 (2003)

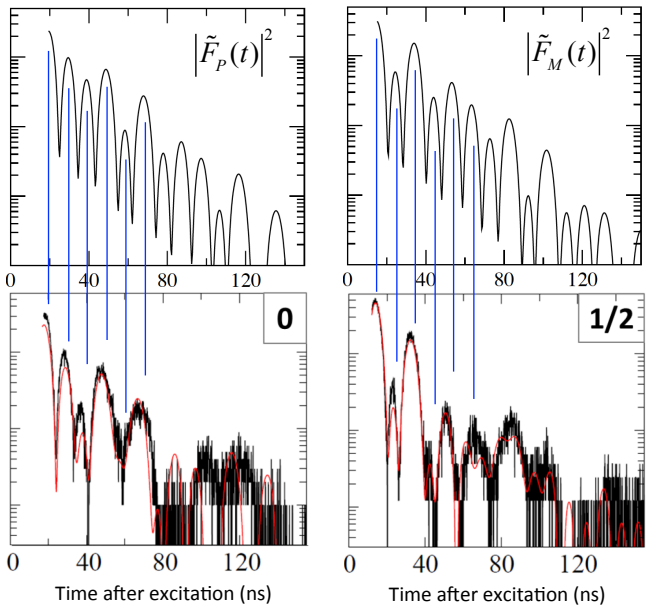

FIG. S2. Comparison of the kinematic description, coherent nuclear resonant scattering simulations, and measurements. The time spectra in the upper row are calculated in the kinematical approximation and provide a qualitatively good agreement with the measured data over the first 70 ns of the measured time spectra. The CONUSS simlation includes the full dynamical theory of nuclear resonant reflectivity. The simulation results (red lines) are plotted along with the measured data and provide a quantitative description of the measurements.