Supplementary Materials for

Imaging mesoscopic antiferromagnetic spin textures in the dilute limit from single-geometry resonant coherent x-ray diffraction

Martin Bluschke et al.

Corresponding author: Martin Bluschke, martin.bluschke@ubc.ca; Alex Frano, afrano@ucsd.edu

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MAGNETIC ORIGIN OF THE $q = (\frac{1}{4} \frac{1}{4} \frac{1}{4})$ DIFFRACTION PEAK

Here we review evidence that the $q = (\frac{1}{4} \frac{1}{4} \frac{1}{4})$ diffraction peak originates from Ni antiferromagnetism, and briefly draw the reader’s attention to the presence of a structural transition in the LSAT substrate near 150 K\textsuperscript{60}. Despite the near coincidence of the substrate structural transition and the PrNiO$_3$ antiferromagnetic transition, the LSAT structural distortion does not give rise to a diffraction peak at $q = (\frac{1}{4} \frac{1}{4} \frac{1}{4})$ in the pseudocubic setting of the PrNiO$_3$ film. Instead, the energy, polarization and temperature dependence of the $q = (\frac{1}{4} \frac{1}{4} \frac{1}{4})$ diffraction peak collectively confirm that the peak is associated with diffraction from a non-collinear antiferromagnetic spin arrangement on the Ni sites of the PrNiO$_3$ film. Fig. S1 shows the incident photon energy dependence of the RXS intensity at $q = (\frac{1}{4} \frac{1}{4} \frac{1}{4})$. The lineshape of the resonant enhancement at the Ni L$_3$ edge matches the expectation for magnetic scattering from the Ni spin lattice (see for example Ref.\textsuperscript{23}). Away from the resonance, the peak is completely undetectable, indicating that the observed Bragg peak originates from scattering in the PrNiO$_3$ film alone.

![Incident Photon Energy Dependence](image)

**FIG. S1.** Ni L$_3$ resonance spectroscopy. Comparison of the x-ray absorption spectroscopy (XAS) intensity measured in the fluorescence yield (FY) mode (black) and the incident photon energy dependence of the resonant x-ray scattering intensity (RXS) at $q = (\frac{1}{4} \frac{1}{4} \frac{1}{4})_{pc}$ (red).

Previously, we characterized the incident-photon polarization dependence of the low-temperature magnetic peak as a function of the azimuthal angle (rotation of the sample about $q = (\frac{1}{4} \frac{1}{4} \frac{1}{4})$). We are able to successfully model the polarization dependence in terms of a period 4 non-collinear spin spiral, with moments $\sim 28^\circ$ from the [0 0 1]
direction in the plane whose normal is [1 − 1 0]. These measurements and the associated model simulation are plotted in Fig. S2, and have also been published in Ref.25. These previous results were obtained on the same film studied in the current manuscript. Although the polarization dependent measurements were performed at low temperature where the antiferromagnetic phase is fully established, we remark that the higher temperature regime in which the small-$q$ modulation is observed is shifted on warming and cooling in agreement with the observed hysteresis of the antiferromagnetic phase, indicating a common origin of both the low and high-temperature signal.

**FIG. S2. Azimuthal dependence of the magnetic peak.** The ratio of scattering intensities for incident $\pi$ and $\sigma$ polarized x-rays is plotted as a function of the azimuthal rotation about $q_{\text{AFM}} = (\frac{1}{4}, \frac{1}{4}, \frac{1}{4})_{pc}$. Experimental data (dark orange) is plotted alongside a simulation of the polarization dependent magnetic scattering from antiferromagnetic Ni spins in the PrNiO$_3$ film (blue). The shaded regions correspond to azimuthal angles for which either the incoming or the outgoing beam is shadowed by the sample horizon in the $\chi = 55^\circ$ wedge geometry. The data point at $\Psi = 90^\circ$ was collected in a $\chi = 0^\circ$ geometry, without the wedge.

**DETECTOR ORIENTATION IN RECIPROCAL SPACE**

The $\chi = 55^\circ$ scattering geometry used to align the momentum transfer along the pseudocubic body diagonal results in a non-trivial projection of the detector plane onto the $H$, $K$, and $L$ reciprocal space directions. This orientation of the detector in reciprocal space is depicted in Fig. S3(a, b). In order to interpret the small-$q$ modulations in terms of interfering diffraction from domains separated laterally in the plane of the film ($a-b$ plane), the diffraction pattern is projected onto the $H-K$ plane. This has been done in Fig 2. After projecting out the $L$ component, the detector direction having had most $L$ component is effectively stretched. In Fig. S3 (d-g) we demonstrate the effect of unstretching the real and reciprocal space images after projecting onto the $a-b$ and $H-K$ planes. In the main manuscript we have chosen to present the reciprocal space patterns and associated real-space domain configurations in their stretched form, thereby emphasizing how the stretching of the $H-K$ plane diffraction pattern onto the skew detector translates into a stretched real-space image following the inverse Fourier transform. The reader should note that it is only possible to project out the $L$ component and directly interpret the modulation patterns on the detector in terms of $H-K$ plane diffraction due to our pre-existing knowledge that the scattering is constrained to the quasi-2D thin film geometry. In our case, the antiferromagnetic domains being probed are pancake-like objects with hundreds of nm correlation lengths in the $a-b$ plane, but constrained in the growth direction by the 40 nm film thickness.

In Fig. 2 of the main manuscript, the depictions of the $q = (\frac{1}{4}, \frac{1}{4}, \frac{1}{4})$ peak and of the model domains in real space give the impression that these are highly anisotropic in the $H-K$ and $a-b$ planes. However, simply by projecting
FIG. S3. Detector orientation and alternate visualizations of the real and reciprocal space patterns. (a) Depiction of the detector oriented in 3-dimensional reciprocal space. (b) Detector in reciprocal space viewed along the $H$ direction. This perspective captures the strong coupling of $K$ and $L$ components along the detector direction perpendicular to $(1\ 0\ 0)$. (g) Experimental diffraction pattern captured by the detector with scale bars in the projected $H - K$ plane ($0.001$ Å$^{-1}$) given by the lengths of the white arrows. (c) Same pattern projected onto the $H - L$ plane and (e) projected onto the $H - K$ plane. The peak elongation appears primarily in the $L$ direction. (f) Domain arrangement in the stretched real space depiction whose squared Fourier transform corresponds to the experimentally observed diffraction pattern in (g). The scale bars in (f) both correspond to $1$ µm along the $[1\ 0\ 0]$ (long bar) and $[0\ 1\ 0]$ (short bar) directions. The stretching of the real and reciprocal space images in (f) and (g) results from projecting out the significant $L$ component along one direction of the detector and then interpreting the pattern in terms of $H - K$ plane diffraction. The unstretched real and reciprocal space patterns (equal $1$ µm scale bars in both directions) are shown in panels (d) and (e). In addition to unstretching, the patterns in (d) and (e) have been rotated to align the $a/b$ and $H/K$ directions with the page horizontal and vertical.

out the $L$ or $K$ components of the detector, as done in Fig. S3(c, e), it is possible to conclude that the peak elongation occurs primarily along the reciprocal space $L$ direction. The associated reduction of the correlation length along $[001]$ is in fact expected due to truncation of the magnetic structure in the film growth direction. The slight asymmetry of the peak in the $H - K$ plane does not have an obvious explanation, though we emphasize that this asymmetry is relatively minor.

**POSITION AND TEMPERATURE DEPENDENCE OF THE SMALL-\textit{q} SUPERSTRUCTURE**

In order to understand the notable variations in the symmetry and wavelength of the small-$\textit{q}$ superstructure observed near the critical temperature in Fig. 1(b) of the main manuscript, we carefully revisited the temperature dependence of these features. Slight temperature changes result in micrometers of cryostat expansion/contraction, which significantly alter the position of the beamspot on the sample and effectively convolute the temperature and position dependence of the observed diffraction patterns. In order to deconvolute the independent effects of temperature and sample position, we performed a detailed characterization of the relative beam-sample motion for a series of temperatures in the vicinity of the transition. Our data reveal that the basic features of the small-$\textit{q}$ modulation depend strongly on sample position, but are essentially independent of temperature across the narrow temperature regime where they are observed. This points to a dominant role of structural defects in determining the antiferromagnetic domain geometries in our film. That is, in the near-critical temperature regime only the few film regions with the highest local $T_{\text{MIT}}$ are ordered, giving rise to a small-$\textit{q}$ superstructure on the antiferromagnetic Bragg peak. As demonstrated in previous studies, the precise value of $T_{\text{MIT}}$ at a given location may depend on structural details$^5$. This interpretation is further supported by the observation that the same location-specific patterns are recurrent after warming to temperatures well into the paramagnetic phase. The experimentally observed RCXD pattern for 3 beam positions on the sample are shown in Fig. S4. The first column shows the diffraction patterns as observed upon warming during a first cycle through the transition. The remaining columns show diffraction patterns observed upon warming during the second
cycle (approximately 24 hours later and after warming to 300 K) and for a series of temperatures in the vicinity of the antiferromagnetic transition. Looking carefully at these diffraction patterns, it is clear that the same basic features of the small-q modulations are retrieved when returning to the same sample position and approximately the same temperature, even after fully cycling through the entire transition. At the lowest temperatures shown, the diffraction patterns begin to change due to the sudden appearance of additional ordered domains within the beamspot.

**FIG. S4.** Position and temperature dependence of the small-q superstructure. Antiferromagnetic RCXD patterns are recorded in the near-critical regime of the antiferromagnetic transition in PrNiO$_3$. Data is presented for 3 non-overlapping sample positions. Each row corresponds to a unique sample position, whereas each column corresponds to a fixed temperature value from either the first or second cycle, as labelled. The second cycle through the transition was performed approx. 24 hours after the first cycle, and after warming to 300 K and cooling to 30 K in between.

As stated in the main text, the specific features of the small-q modulations seen in Figs. 2(d,i) are altered as soon as the sample is translated with respect to the beam by approximately 1 beam width (10 $\mu$m). This observation rules out an extended periodic real space domain arrangement with the same symmetries as the model domain arrangements shown in Figs. 2(a,b,f,g). To further confirm this picture, in which the ordered domains are dilute in the beamspot, we have integrated the peak intensity as observed on the CCD for a series of sample positions (step size 2 $\mu$m). The resulting intensity-position profile is plotted in Fig. S5 and demonstrates regions of both high and low intensity, with many of the high intensity regions having a width approximately equal to the beam size. The latter observation is indicative of isolated domain arrangements whose total spatial extent is small compared to the beam width, in agreement with the result of our modelling. Such regions are highlighted by the presence of a horizontal black bar whose length corresponds to the 10 $\mu$m beam width. The two curves plotted in Fig. S5 represent equivalent measurements taken during the first and second cycles at comparable temperatures. The similarity of the profiles observed during the 2 cycles further indicates that the spatial inhomogeneity of the antiferromagnetic order parameter is associated with a corresponding structural inhomogeneity, which is insensitive to the temperature cycling.

**BCDI SAMPLING REQUIREMENTS**

In order to perform a Bragg coherent diffractive imaging (BCDI) type experiment, a series of technical requirements must be fulfilled so as to satisfy the so-called Nyquist/Shannon sampling criteria$^{20}$. Loosely speaking, these criteria imply that the angular resolution of our CDD detector (solid angle covered by 1 pixel) should be, at most, half the width of one speckle. Alternately stated, the minimum speckle size must be 2 pixels in order to ensure a sampling sufficient for reconstructing the shape of the scatterer(s) in real space. Whether or not these criteria are fulfilled depends on a combination of the probe photon wavelength, the beam size (or the sample support, whichever is smaller), the sample-detector distance, and the detector pixel size (for a complete discussion see Ref. $^{29}$). In our experiment, the sample-detector distance and the CCD pixel size were fixed by the experimental infrastructure present at the CXS.
beamline. Furthermore, the photon wavelength is fixed due to the necessity of working at the Ni L₃ resonance. For these experimental conditions the largest beam size which satisfies the sampling requirements is 8.2 µm. Although we used a 10 µm beam spot, the effective size of the scattered beam was significantly smaller, being defined by the spatial extent (< 5 µm) of the ordered domains in Figs. 2(a, b, f, g), which were dilute in the beamspot in the investigated temperature regime. In this sense, the system we studied is “self-constraining” in the dilute regime. Our results demonstrate that with a self-constraining sample it is possible to do BCDI despite not having perfect transverse coherence and despite having too large of a beam for the Nyquist/Shannon sampling.

FIG. S5. Magnetic peak intensity as a function of beam position. The integrated antiferromagnetic Bragg peak intensity recorded on the CCD is plotted as a function of the relative beam-sample position. The two curves were taken upon warming during separate cycles through the antiferromagnetic-paramagnetic transition. The black bars indicate the width of the beam (10 µm), and highlight features whose width corresponds approximately to one beam width. The sample position scale has been given an arbitrary starting point.