FIRST OBSERVATION OF A MAGNETIC SPECKLE PATTERN BY COHERENT X-RAY SCATTERING AT THE URANIUM M_{IV} EDGE

F. YAKHOU[1], A. LÉTOUBLON[2], F. LIVET[2], M. DE BOISSIEU[2], F. BLEY[2] AND C. VETTIER[1]

1 ESRF, EXPERIMENTS DIVISION
2 LTPCM-ENSEEG, UMR-CNRS/INPG/UJF N°5614, ST MARTIN D'HÈRES (FRANCE)

The availability of high fluxes of coherent hard x-rays has opened up new fields of investigations on disordered systems, through the analysis of the random diffraction or "speckle" patterns. A promising application is the study of the disorder inherent to domain formation in magnetic systems.

Taking advantage of the high quality focusing optics of the ID20 beamline and the huge enhancement of the magnetic scattering amplitude when the energy is tuned to the M-edges of uranium (< 4 keV), we were able to record a magnetic speckle pattern from the antiferromagnetic phase of a UAs sample.

When scattered by coherent incident radiation, any disordered material introduces random phase-shifts that result in a strong modulation of the average diffraction pattern yielding the graininess characteristic of a speckle pattern. Each such pattern is related to the exact spatial arrangement of the disorder and to the coherence properties of the incident radiation. It will evolve together with both these features, making the knowledge of the properties of the incident radiation essential to retrieve meaningful information on the sample itself.

Extracting the exact arrangement of the sample from a static speckle pattern, though theoretically possible, is in practice a daunting task. Much of our experimental work has been dedicated to intensity fluctuation spectroscopy by analyzing the time correlation of the intensity on a single point of a speckle pattern, thus giving information on the underlying dynamics with nearly atomic resolution and frequencies ranging in practice from 10^{-3} Hz to 10 Hz.

Antiphase domain structure in Cu_3Au, equilibrium critical fluctuations in Fe_3Al, brownian motion of gold colloids in glycerol, dynamics of block copolymer micelles have been successfully addressed by coherent scattering techniques in the 8 keV energy range [1], where a high flux of coherent x-rays can be "easily" obtained. A new exciting domain of applications will be the study of magnetic domains.

Because of crystal symmetry, domain formation is inherent to most types of magnetic ordering but little is known up to now on domain size and arrangement. A speckle pattern from a magnetic system should prove of considerable interest in this respect. Since the main difficulty lies in the extreme weakness of the magnetic signal, UAs was chosen as a test system for which advantage can be taken from the huge enhancement of the magnetic intensity through a resonant process at the uranium M_{IV} absorption edge (3.73 keV). The unavoidable counterpart is a lower flux at the sample due to higher absorption by windows. UAs has the crystal structure of NaCl with a lattice parameter $a_0 = 5.766$ Å and orders at $T_N = 127$ K in a type-I antiferromagnetic structure with alternating ferromagnetic sheets stacked along the c-axis.

A 20 μm collimating pinhole selected the coherent fraction of a 3.73 keV beam from the first harmonic of two phased 42 mm period undulators, its spectral width given by the 1.3 x 10^{-4} bandwidth of a Si (111) double crystal monochromator. A secondary set of slits of adjustable aperture placed after the optics, 3 meters from the pinhole, acted as the effective source, thus avoiding any spoiling of the coherence by optical elements. The second crystal of the monochromator and the second mirror were used to focus the beam horizontally and vertically respectively, down to 350 x 80 μm^2 in the pinhole plane. The resulting integrated flux through the 20 μm pinhole was as high as 7.10^7 ph/s at 180 mA ring current with a 60 x 60 μm^2 opening of the secondary

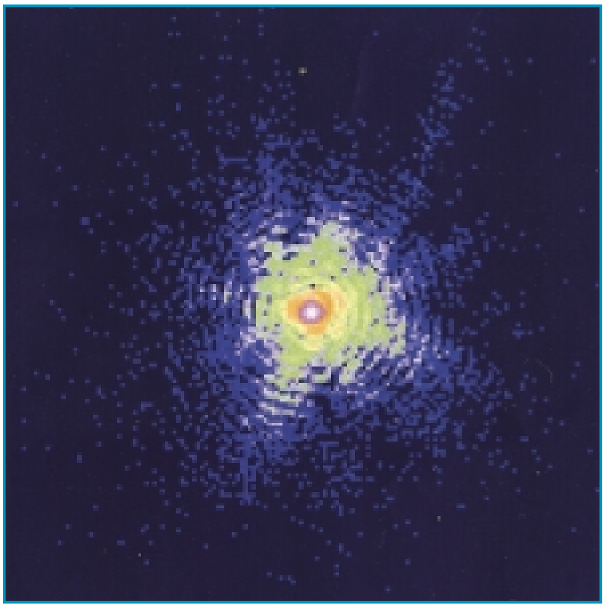

***Fig. 1:** Fraunhofer diffraction pattern from a 10 µm pinhole at 3.73 keV. The fringes are visible up to the tenth order, indicating the high degree of coherence of the illuminating radiation.*

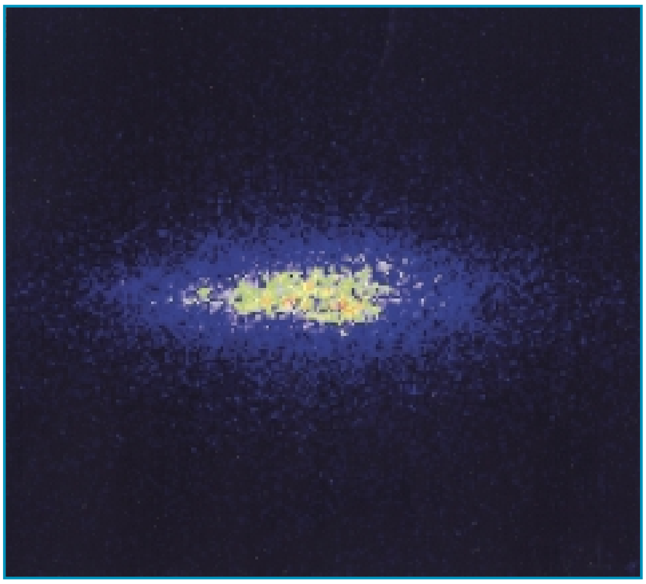

Fig. 2: Speckle pattern from magnetic domains in the type-I antiferromagnetic phase of UAs, at the uranium M_{IV} resonance (3.73 keV). Horizontal and vertical cuts are displayed in Fig. 3.

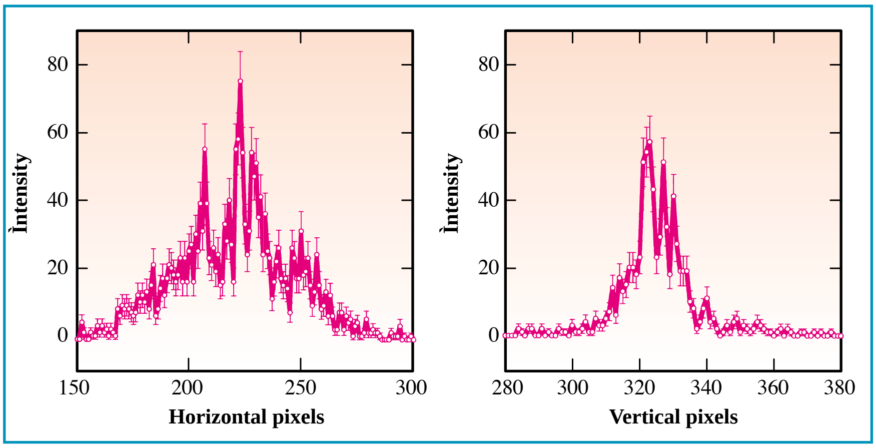

Fig. 3: Horizontal and vertical cuts through the speckle pattern of Fig. 2. The error bars correspond to Poisson statistics and cannot account for the raggedness in the scattering.

slits. It is worthwhile noting that we could obtain up to 7.10^8 ph/s at 140 mA current through a 10 μm pinhole and 60 x 60 μm^2 slits at 7.6 keV, with a 48% degree of coherence (ß) at small angle, which is by far the highest and best coherent flux achieved up to now.

Both a standard Bicron scintillation detector and a Princeton Instruments direct illumination CCD chip with 384 x 576 22 μm square pixels could be used as detectors, 1.8 m away from the sample. The CCD camera was used as a 2D photon counter by applying a droplet algorithm [2] that could identify individually each photon reaching the camera.

The coherence properties of the beam were retrieved from the statistical analysis of the static speckle pattern produced by a silica gel following an earlier paper [2]. ß, as given by the contrast of the speckle pattern, was found to be around 50%, though evolving with time down to 30% within half an hour, indicating instabilities in the experimental set-up that we suspect to come from temperature variations in the experimental hutch. The Fraunhofer diffraction pattern of a 10 μm pinhole at 3.73 keV is displayed on Figure 1. The good contrast of the fringes up to the tenth order is a clear sign of the high coherence of the illuminating beam.

The 5 x 5 x 2 mm^3 UAs sample was oriented with a [001] cleaved surface and put in a cryostat with 2 x 500 μm thick inner Be windows and 2 x 125 μm thick outer Kapton windows, giving a total 2% transmission at 3.73 keV. Because of practical limitations in the detector 2θ angle, the crystal orientation and intensity optimization were done at 3.75 keV. The sample was then cooled down to 100 K, in the magnetically ordered phase. By tuning the energy to the maximum of the resonance we achieved 60 counts per second at peak maximum on the (001) antiferromagnetic reflection from a ≈ 40 μm single grain, a result somewhat expected from previous non-coherent experiments on the very same sample. A time-series of 181 summed up 10 second each acquisition frames is represented on Figure 2. A speckle structure is clearly visible. Vertical and horizontal cuts through the image are displayed in Figure 3, with error bars corresponding to Poisson counting statistics that clearly cannot account for the raggedness in the scattering.

Because of crystal symmetry, one would not expect the diffraction pattern to be anisotropic. The apparent 1:≈4 anisotropy of the scattering can be accounted for by a considerable smearing of the (001) reciprocal lattice node along the scattering vector due to strong absorption effects. For the given experimental geometry the 2D CCD image can be viewed as a planar cut in the reciprocal space perpendicular to the diffracted wavevector, thus at an angle $\theta_{001} = 15°$ to the scattering vector. That would result in a cosθ:sinθ anisotropy of the diffraction pattern.

Using the high coherent flux at 3.73 keV that was obtained on the ID20 beamline, we have demonstrated the feasibility of coherent scattering experiments on magnetic systems that should give a unique insight on the physics of magnetic domains. The even higher flux obtained at 7.6 keV should allow investigations at the L absorption edges of rare-earth compounds with weaker resonances, opening up this new field to a wide variety of magnetic systems. ■

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