Source: Alastair Rae

Melting the antiferromagnetic ice sheet

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Polar bears and physicist share a strong interest in phase transitions. Polar bears take the real world irregularities that cause their ice flow to break up in fixed frozen and more dynamic molten parts rather stoically. For physicists however, who like to think in terms of ideal ordered systems, real world disorder is just bad news. Pet theories like critical scaling, which describe the divergence of the size of fluctuations at phase transitions, break down in the presence of disorder. Take for instance thin films, where structural disorder can never be avoided. As a result, magnetic phase transitions in such films are difficult to understand, a situation that is quite disturbing in view of the importance of such films in science and technology.

To oversee the battle between order and fluctuations in phase transitions and to understand the role of structural disorder in this fight requires a tool that covers a huge range of length- and timescales. While the length scale of fluctuations can be readily probed by conventional diffraction experiments, the observation of the corresponding fluctuation times is much more challenging. As we will show here, coherent soft X-rays turn out to provide a remarkably powerful looking glass for this purpose.

As a test case, we studied an epitaxially-grown ultrathin 11-monolayer holmium film displaying a helical magnetic order with a periodicity along the surface normal [1]. This order disappears in a broad transition around T_N=76 K. In the experiment the coherence of the light of the beamline (U49-2 PGM 1 and UE46 PGM) is obtained by a spatial filter consisting of two pinholes (Fig. 1). The periodic helical magnetic structure produces strong magnetic diffraction peaks, when the photon energy is tuned to the M_5 resonance of holmium. One these peaks we intercept with a CCD camera. In this way we can observe the melting of the magnetization, much like polar bears watch the ice melt under their feet. Unlike the polar bears however, we can regulate the temperature in our experiment within 10 mK and in this way control the magnetic correlation length ξ, the typical length scale over which the sample is magnetically ordered. With incoherent light we would observe one smooth diffraction peak, with a width that is inversely proportional to the correlation length of the ordered regions. As expected, we find that the closer we approach the transition temperature T_N, the smaller is the scattered intensity (Fig. 2). The correlation length also decreases, although in a way that is quite different from standard models. What is happening here? In order to find out we make our light coherent and study the fluctuations in the time domain.

When coherent light is used, the magnetic peak breaks up in a myriad of speckles, which form the diffraction pattern of the magnetic structure of the illuminated spot. Any changes in this structure are immediately reflected in changes in the speckle pattern. By recording the speckle-pattern as a function of time and performing a time correlation analysis of the speckle intensities it is possible to extract the correlation time of the fluctuations.

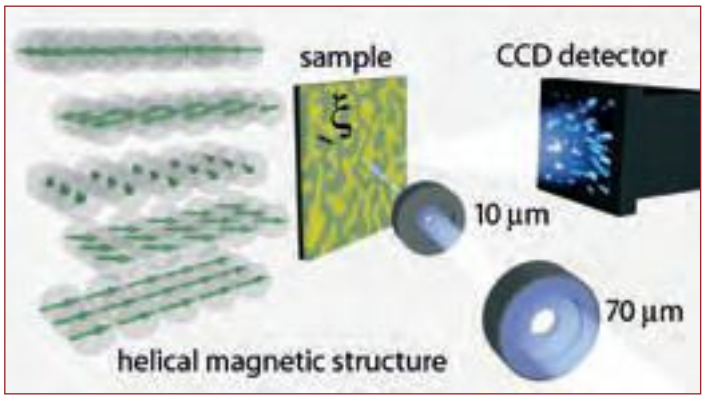

Fig. 1:
Sketch of the coherent diffraction experiment and the helical magnetic structure of holmium. The length scale over which the magnetic structure is ordered in the plane of the sample is depicted by the correlation length ξ.

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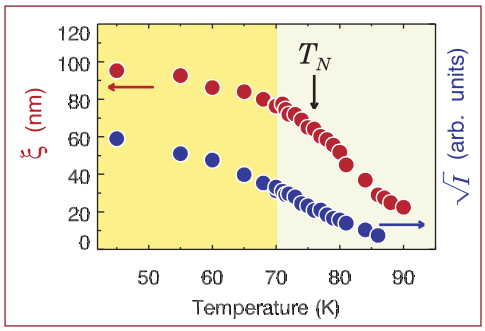

Fig. 2:
Temperature dependence of the correlation length ξ and intensity I. The fluctuations could be measured in the time domain up to 70 K.

This technique is called Photon Correlation Spectroscopy [2] which is well known from laser light scattering where, among other things, it is used to study diffusion in colloidal systems. With the availability of brilliant X-ray sources, this laser technique has started to make inroads into the X-ray regime [3], where the fluctuations can be measured to much smaller correlation lengths. So far, only soft condensed matter systems have been probed such as colloids [4] or smectic films [5].

Here we use the technique to follow correlations in a hard condensed matter system, namely the magnetic fluctuations in our film. Fig. 3 shows one of the speckle patterns with an exposure time of 4 seconds together with snapshots of the 3-hour speckle movies, some of which can be viewed online [6]. From these it is clear that at 50 K the magnetic structure is fixed, but that at higher temperatures fluctuations appear that speed up dramatically.

Moreover, when all images in a movie taken at a fixed temperature are averaged, we find something remarkable. At 50 K, the intensity profile has a contrast close to a single speckle pattern of 4 s exposure, confirming a frozen domain structure (Fig. 4). At 70 K the intensity profile blurs out, but still some fixed speckles remain. This proves that the magnetic fluctuations are highly constrained by the defect structure of the film. In more technical terms, the system is said to be non-ergodic.

Although there are some important differences, this transition is analogous to the melting of ice, in which the correlation length of the ordered ice phase (e.g. the ice floes) will be reduced when the temperature increases. The limited coherent flux binds us at present to the ultra slow regime, between 4 seconds and 3 hours. From an intensity correlation analysis we find that this time regime corresponds to the fluctuation times in our sample at temperatures below roughly 70 K.

This is the first time such magnetic correlations have been followed not only in the space but simultaneously in the time domain. The movies undoubtedly show that the strange behavior of the correlation length is not only due to the reduced dimensionality of the film, but also due to real world defects. The challenge now is to determine the temperature-dependent ratio of the dynamic to static parts of the sample in this magnetic system. Meanwhile beckoning on the horizon are the plethora of correlated electron systems showing stripes, charge and spin density waves or other long range order effects which may or may not be dynamic in nature. With the 1,000 times higher coherent flux that can be expected at optimized beamlines [7] or the 10^9 times higher flux provided by X-FELs, the future for this technique looks excellent.

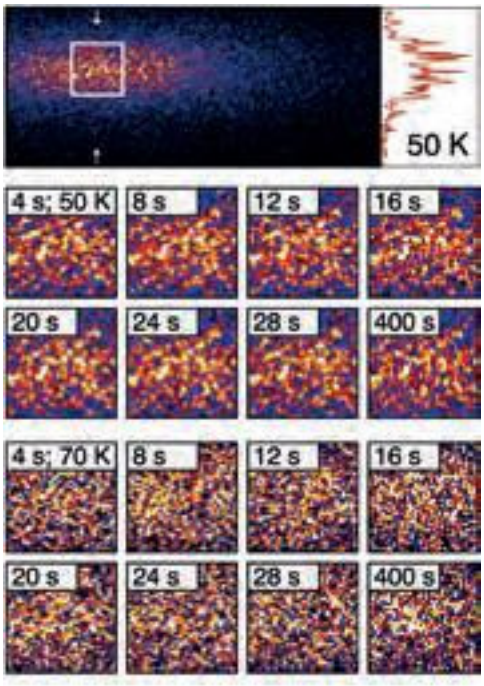

Fig. 3:
Top: speckle pattern at 50 K. Below: examples of 4 s exposures acquired at the indicated time in the area marked in the top panel at 50 K (top rows) and at 70 K (bottom rows), showing the fluctuations in the speckle pattern that reflect the fluctuations in the magnetic structure. At 50 K the speckle pattern is almost completely static but is fluctuating rapidly at 70 K (6 K below the film's ordering temperature).

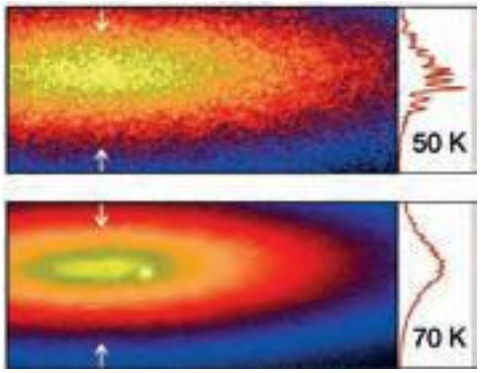

Fig. 4:
Intensity profile obtained by averaging over 2,200 speckle patterns acquired over about three hours.

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