

Imaging Antiferromagnets using Second Harmonic Generation

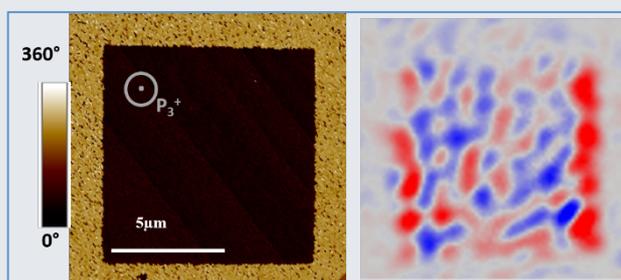
M. Viret, J.-Y. Chauleau, T. Chirac (SPEC, CEA), S. Fusil and C. Carétéro (UMPhy, CNRS/Thales).

Spintronic devices are largely based on ferromagnetic metals (FM) in multilayer stacks. So far, antiferromagnets (AF) are only useful indirectly, because of their coupling which pins the magnetic moments of FM layers. The realization that spin currents can produce a magnetic torque on the AF order opens a new route towards introducing these materials as active elements in spintronics.

In CEA/SPEC, we have started a new activity aiming at opening the fields of spintronics and magnonics to antiferromagnetic (AF) insulators. Our goal is to demonstrate that pure spin currents, strain and/or femtosecond light pulses can be used to manipulate the AF order in some well-chosen AF insulators. Imaging techniques are therefore essential because one needs to visualize AF domains to assess the various writing parameters. Because of the absence of stray fields in these materials, imaging is a very challenging task. Presently, the most efficient techniques rely on using linearly polarized light (synchrotron or visible) to measure the direction of the AF vector. However, another technique based on non-linear optics can also be very powerful. Indeed, when illuminated by intense light, some non-linear materials can re-emit higher harmonics. This is allowed in non-centrosymmetric materials as well as on surface and interface states of centrosymmetric media owing to the breaking of inversion symmetry at the boundaries. Similarly, the breaking of time-inversion symmetry by long-range magnetic ordering leads to new contributions to SHG. As these depend on the direction of AF vectors, they can be used to probe the magnetic structure.

We have recently setup such a high-resolution SHG imaging in transmission mode in our laboratory. It uses laser pulses emitted from an amplified Ti-Sapphire laser whose wavelength can be continuously adjusted by Optical Parametric Amplification from 2600 nm to 530 nm. The emitted light impinging on the sample is linearly polarized and can be rotated using a $\lambda/2$ prism. Right after it, a custom-made interference filter completely rejects the first harmonic light, before the second harmonic is collected by a high numerical aperture objective and is imaged by a sensitive Peltier cooled CCD camera (acquired through the labex).

Typical antiferromagnets like NiO have typically 12 kinds of AF domains, which makes any attempt to disentangle magnetic contrasts a difficult challenge. This situation can be largely simplified in multiferroics, materials where another order can couple to antiferromagnetism. Indeed, by writing a single ferroelectric domain in the archetype BiFeO₃, the number of AF variants is reduced to three, making a quantitative analysis feasible. We have recently achieved to image both ferroelectric and AF domains in a BFO(50nm)/SrRuO₃/SrTiO₃(001) thin film electrically written using a Piezo-Force microscope. The AF domains reconstructed in a single polarization pad, after a full polarization analysis of the light emitted in every pixel, have typical micron sizes (see Figure). This allows to test several external stimuli to attempt writing AF domains, thus paving the way to using insulating antiferromagnets as memory elements.



(left) PFM image of a PFM written ferroelectric monodomain in BiFeO₃. (Right) SHG reconstructed AF contrast showing AF domain in the micron range

Importantly, the technique is compatible with time-resolved imaging using pump-probe methods. It consists in sending a pump laser pulse and imaging with a probe pulse arriving with some time delay. The time evolution of the AF domains is thus achievable at a time-scale of the order of the pulse duration, i.e. 100 fs. This should allow us in the near future to probe antiferromagnetic dynamics, which is expected in the THz range.

J.-Y. Chauleau, E. Haltz, C. Carrétéro, S. Fusil and M. Viret, *Multi-stimuli manipulation of antiferromagnetic domains assessed by second harmonic imaging*, Nature Mat. 16, 803 (2017)

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