Circularly Polarized X-ray Magnetic Scattering as a probe of Multiferroic Materials

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Multiferroic materials (in which electric and magnetic order coexist) that also display magneto-electric (ME) coupling have raised considerable interest in recent years, as candidate compounds for the development of advanced applications in electronics and data storage. In particular, the ME coupling has shown to be relevant in those materials in which the onset of a non-collinear antiferromagnetic order drives the formation of a ferroelectric state [1]. Of fundamental importance to fully understand the coupling is an accurate microscopic description of the magnetic order.
$TbMnO_3$ is the canonical magnetoelectric multiferroic, in which the phase transition to a cycloidal spin arrangement at T=27 K is concomitant with the onset of electric polarization [2,3]. More recently, $Ni_3V_2O_8$ has also been established as a magnetoelectric multiferroic. Its structure is characterized by a buckled kagome staircase, resulting in two inequivalent Ni sites; below T=6.3 K, the Ni^{2+} S=1 spins are reported to be arranged in a cycloid [4].
A key feature in this class of compounds is the possibility to control the population of magnetic domains (defined by the handedness of the cycloids) by means of an in-situ electric field, directly exploiting the ME coupling.

The combination of magnetic non-resonant diffraction by circularly polarized X-rays with the full linear polarimetry of the scattered beam opens the way to a new class of experiments, in which the magnetic state of complex magnetic materials can be probed, while the magnetoelectric domain formation can be controlled by applied electric and magnetic fields.
This technique brings a strong experimental sensitivity to the imbalance in the magnetic domain populations, since the handedness of the circular polarization naturally couples to the sense of rotation of the magnetic moments, leading to an accurate description of the domain state and to the refinement of the magnetic structure.

The results allowed us to shed more light on the complex magnetic structure of multiferroic $TbMnO_3$, a challenging test case due to its two magnetic sublattices on the Tb and Mn sites. The method led to the identification of components of the ordering on the Tb sublattice and phase shifts that earlier neutron experiments could not resolve [5].
In the case of $Ni_3V_2O_8$, the polarization analysis has been used not only to the refinement of the magnetic structure, but also to obtain real space images of the magnetic cycloidal domains. Their evolution is followed as they are controlled via ME coupling by the applied electric field and cycled through a hysteresis loop, thus collecting valuable information on domain formation, inhomogeneities and domain wall movement [6].

References

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