Birefringence of X-rays

Polarization phenomena in photon scattering, from optics to X-rays, provide information on electronic degrees of freedom not available with any other technique in materials science. In the past decade, striking progress has been made with the analysis of linear and circular polarization data obtained using the highly collimated and tunable X-Ray beams available at synchrotron sources. This is especially the case in resonant Bragg diffraction, where the intensities of weak or forbidden Bragg diffraction spots are greatly enhanced when the energy of the X-ray photon is tuned to a resonance of a selected atom. Unique and valuable information can then be obtained about the spin and orbital properties of the electrons that participate in, for example, magnetism, superconductivity, and multiferroic behaviour. However, a proper interpretation of polarization data requires an accurate theory of the radiation-matter interaction near an X-Ray resonance. This theory must include, in particular, the effects of birefringence.

Below 213 K, Copper Oxide CuO is an antiferromagnet. In a recently reported experiment (V. Scagnoli et al. *Science*, 332, 696 (2011)), the magnetic reflection (i.e. a scattering by the electron spins) was recorded at 100 K for various states of the incoming polarization and with analysis of the outgoing polarization. This was done for photon energies around the Cu L2 and L3 edges at 932 and 953 keV (which correspond to excitations of a 2p core electron to the 3d outer orbitals of the Cu atom). Conventional magnetic diffraction rotates a linear polarized incident beam 90 degrees. Detection of a non-rotated component in the outgoing polarization as well, guided the above authors to the exciting possibility of a scattering from an orbital or "toroidal" electronic current around the copper atoms. This supported a recent suggestion that such multipoles play an important role in High Temperature Superconductivity in copper oxide compounds.

However, since birefringence is permitted in a CuO crystal because of its low symmetry (monoclinic), calculations of the radiation-matter interaction from first principles are required to get a reliable quantitative description of the X-Ray scattering signals, and determine what the dominant physics is. We have done such calculations and, comparing them with the diffraction data kindly supplied by the above authors, we have demonstrated that a better explanation for these experimental results is to be found in birefringence.

We indeed confirmed that the atomic magnetic scattering gives only a 90 degree rotation of the polarization. But we found that, as the beam propagates through the crystal the polarisation rotates significantly (see Fig. 1). This leads to an apparently unrotated polarization component in the finally emerging wave. We have been able to reproduce quantitatively all the experimental results purely in terms of birefringeance: the spectrum shape, its polarization dependence and its azimuthal variation (i.e. the variation with rotation of the sample around the normal to the diffracting plane). To complete our analysis, we have also performed simulations that include the scattering process related to the hypothetical toroidal current in the material. We always found its contribution to be extremely small.

This may not necessarily mean that the orbital current idea is a false trail in the quest to understand High Temperature Superconductivity, other techniques are required to elucidate that. Nevertheless, this case has demonstrated how birefringence can play a major role in X-Ray resonant diffraction experiments and that precise simulations are essential to correctly interpret energy spectra and azimuthal and polarization dependences in such experiments.

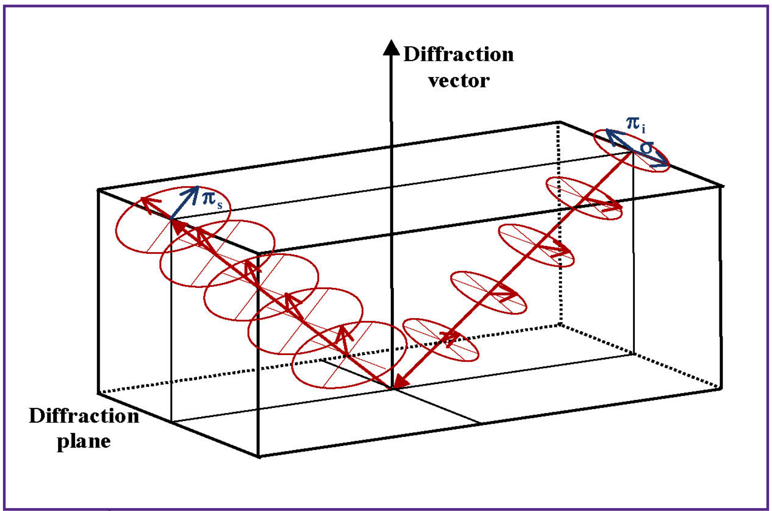

Fig.1: As the electromagnetic wave propagates in the birefringeant material, its polarization rotates progressively. When magnetic scattering occurs from a specific diffraction plane at some depth in the material, the polarization rotates by 90°. The outgoing wave then continues its slight rotation up to exit from the crystal. Because of the birefringeance, the observer measures an unexpected polarization state, a rotation different from 90 degrees.

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FURTHER READING

BIREFRINGENCE AND POLARIZATION ROTATION IN RESONANT X-RAY DIFFRACTION
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