

Atomic-Plane Resolved Electron Magnetic Circular Dichroism by Achromatic Spatially-Resolved Electron Energy Loss Spectroscopy

Xiaoyan Zhong¹

¹ National Center for Electron Microscopy in Beijing, Key Laboratory of Advanced Materials (MOE), The State Key Laboratory of New Ceramics and Fine Processing, School of Materials Science and Engineering, Tsinghua University, Beijing, P.R. China.

The atomic-level knowledge of local spin configuration of the magnetic materials is of great importance to predict and control their physical properties, in order to meet the challenges of ever-increasing demands on performance of functional materials. However, it is highly challenging to experimentally characterize magnetic properties of such materials with atomic scale spatial resolution.

The leading techniques in spatially resolved magnetic imaging are magnetic exchange force microscopy and spin polarized scanning tunneling microscopy. However, as they are surface sensitive, very little information can be obtained regarding bulk or buried materials. The X-ray magnetic circular dichroism (XMCD) combined with photoelectron emission microscopy technique is a very attractive alternative because it has the spatial resolution as high as the polarized x-ray beam size besides element specific feature, as it is less surface sensitive and can be used to look at the interior of the thin films. However, the length scale of magnetic contrast using highly brilliant left and right circularly polarized X-ray beams is around 15nm [1].

The best option to push the spatial resolution of the spectromicroscopies lies in the electron beam equivalent technique electron energy-loss magnetic chiral dichroism (EMCD) [2], which is also called electron magnetic circular dichroism. Physically, XMCD and EMCD shares the same underlying physics in which the angular momentum transferred during X-ray absorption or inelastic electron scattering can selectively excite magnetic sublevels in atoms. The structured electron beams generated through interference of suitably phased plane waves can produce beams with orbital angular momentum. Electron beams can be easily focused compared with X-rays, allowing for atomic scale magnetism to be probed. Previously, we have found a strong EMCD signal in transition metal oxides allowing them to use standing wave methods to identify the different spin states of Fe atoms with site specificity [3].

In principle EMCD can offer higher spatial resolution and greater depth sensitivity due to the short de Broglie wavelength and penetration of high-energy electrons compared to XMCD. Our approach combines spatially-resolved EMCD with the latest developments in chromatic aberration corrected electron microscopy, which reduces the focal spread of inelastically scattered electrons by orders of magnitude when compared with the use of spherical aberration correction alone [4]. In the example of complex oxide of ferromagnetic $\text{Sr}_2\text{FeMoO}_6$ with a Curie temperature of ~ 420 K and a tetragonal double-perovskite structure [4,5], magnetic circular dichroism spectra of Fe are imaged atomic plane by atomic plane as shown in Figure 1, which can provide quantitative information of element-selective orbital and spin magnetic moments at the atomic level. Combining with advanced capability of structural and chemical imaging by using aberration-corrected transmission electron microscopy, all the information including magnetic polarization, atomic configurations, and chemical states can be simultaneously accessed from the very same sample region [6-8].

The spatial resolution of atomic-plane resolved EMCD method goes beyond that of any currently available technique, including XMCD and neutron diffraction. The structural, compositional and bonding information can be correlated with local spin configurations on the atomic scale, providing deep insight into structure-property relationships in magnetic materials at the atomic level. These information are not only contribute to a fundamental understanding of the local interplay between charge, spin, orbital and lattice degrees of freedom in magnetic functional materials, but also pave the way for new designs of magnetic materials for future applications with improved device functionality [9].

References:

- [1] WL Chao et al, *Nature* **435** (2005), p. 1210.
 [2] P Schattschneider et al., *Nature* **441** (2006), p. 486.
 [3] ZQ Wang et al., *Nature Communications* **4** (2013), 1395.
 [4] ZC Wang et al., *Nature Materials* **17** (2018), p. 221.
 [5] XF Chen, ZC Wang and XY Zhong, *Journal of Chinese Electron Microscopy Society*, **37** (2018), p. 548.
 [6] PL Ho et al., *Ultramicroscopy* **193** (2018), p. 137.
 [7] L Jin et al., *Advanced Materials Interfaces* **3** (2016), #1600414.
 [8] J Lin et al., *Physical Review Materials*, **1** (2017), 071404(R).
 [9] The author acknowledges funding from National Key Research and Development Program (2016YFB0700402), National Natural Science Foundation of China (11834009, 51761135131, 51822105, 51671112, 51788104), National Basic Research Program of China (2015CB921700) and Fund of Key Laboratory of Advanced Materials of Ministry of Education (2018AML12). This work made use of the resources of the National Center for Electron Microscopy in Beijing. Mr. ZC Wang, Dr. AH Tavabi, Dr. L Jin, Dr. J Ruzs, Dr. D Tyutyunnikov, Mr. HB Jiang, Prof. Y Moritomo, Prof. J Mayer, Prof. RE Dunin-Borkowski, Prof. R Yu and Prof. J Zhu are thanked for their valuable contributions to this work.

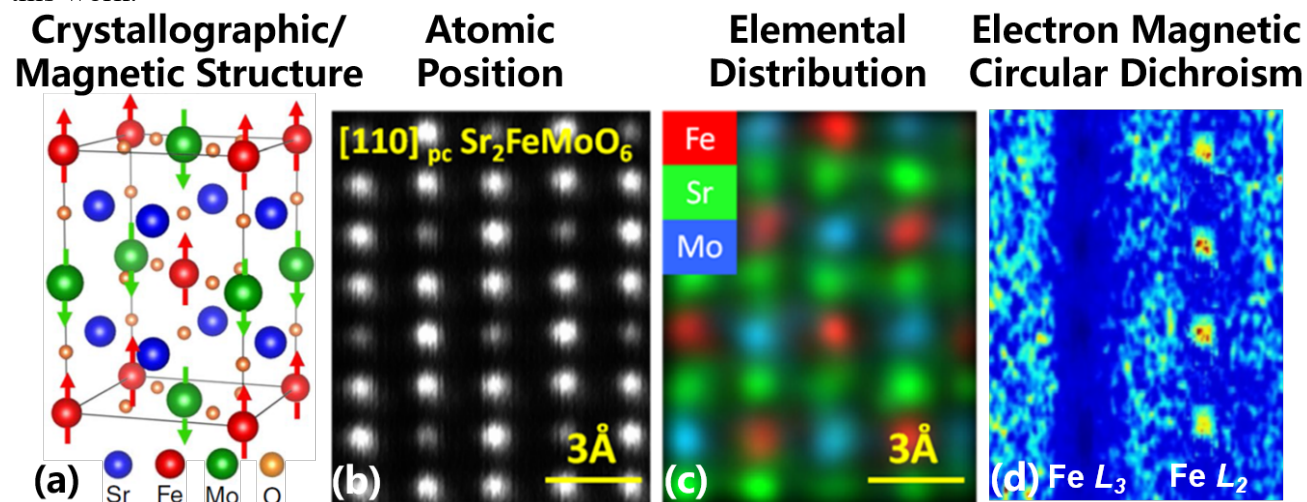


Figure 1. (a) Crystallographic and magnetic structure of ferromagnetic double-perovskite $\text{Sr}_2\text{FeMoO}_6$.^[5] (b) High-angle annular dark field scanning transmission electron microscopy image of $\text{Sr}_2\text{FeMoO}_6$ along a pseudo-cubic [110] zone axis reveals the positions of Sr, Fe and Mo atoms. (c) Atomic scale energy-dispersive spectroscopy mapping shows each columns of Sr atoms in green, Fe atoms in red and Mo atoms in blue. (d) Atomic plane resolved electron magnetic circular dichroism of Fe in $\text{Sr}_2\text{FeMoO}_6$.^[4]