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Detection of electron magnetic circular dichroism signals under zone axial diffraction geometry

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ABSTRACT

EMCD (electron magnetic circular dichroism) technique provides us a new opportunity to explore magnetic properties in the transmission electron microscope. However, specific diffraction geometry is the major limitation. Only the two-beam and three-beam case are demonstrated in the experiments until now. Here, we present the more general case of zone axial (ZA) diffraction geometry through which the EMCD signals can be detected even with the very strong sensitivity to dynamical diffraction conditions. Our detailed calculations and well-controlled diffraction conditions lead to experiments in agreement with theory. The effect of dynamical diffraction conditions on EMCD signals are discussed both in theory and experiments. Moreover, with the detailed analysis of dynamical diffraction effects, we experimentally obtain the separate EMCD signals for each crystallographic site in $Y_3Fe_5O_{12}$, which is also applicable for other materials and cannot be achieved by site-specific EMCD and XMCD technique directly. Our work extends application of more general diffraction geometries and will further promote the development of EMCD technique.

1. Introduction

Since it was demonstrated by Schattschneider et al. [1] in 2006 that electron magnetic circular dichroism (EMCD) signal can be detected in the transmission electron microscope (TEM), different experimental EMCD diffraction geometries have been proposed for signal acquisition to improve the spatial resolution and signal-noise-ratio (SNR) in the last few years, which include the convergent beam electron diffraction (CBED) [2,3], large angle convergent beam electron diffraction (LACBED) [4,5], energy filtered transmission electron microscopy (EFTEM) [6], q–E diagram [7], and scanning transmission electron microscopy (STEM) [8]. Although all of them have their own advantages, they are still totally based on the two-beam or three-beam diffraction geometry to reduce the complexity of dynamical diffraction effects as much as possible in the periodic crystal structure. The two-beam or three-beam case are always based on the systematical reflection conditions, which is also called planar channeling conditions by John Spence in the ALCHEMI technique (Atom Location by Channeling Enhanced Microanalysis) [9]. Tilting the sample to such cases will reduce the number of Bragg spots in the diffraction plane and thus reduce the number of reflections significantly contributing to the total signal. Then the quantitative relationship between EMCD and dynamical diffraction effects can be simplified. In fact, the diffraction geometry for EMCD acquisition is not limited to the case of systematical reflections. A more general case should be under zone axis (axial channeling conditions) with more diffracted beams interfering with each other to finally contribute to the EMCD signals in the reciprocal space.

Intuitively, performing the EMCD measurement with incoming beam parallel to the zone axis will lead to strong dynamical effects as a larger number of diffracted beams appears in the diffraction plane. The distribution of signals in the diffraction plane will become more complex, and can only be described by theoretical simulations with heavy computational efforts due to a need to consider a large amount of diffracted beams. In addition, the calculation results of bcc Fe under the [001] zone axis [10,17] have shown that the magnetic signal forms a complex pattern with many local minima and maxima, changing sign throughout the diffraction plane due to pronounced dynamical diffraction effects. Therefore, strict control of experimental conditions is of great importance for detecting EMCD signal with reliability and high SNR. All in all, the complicated dynamical diffraction effects lead to

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the fact that there are still no experimental results for EMCD technique under zone axial diffraction geometry.

However, the zone axial diffraction geometry has some advantages compared to the two-beam or three-beam case. Firstly, the extreme complexity of dynamical diffraction effects lead to a very high sensitivity of EMCD signals to slight changes of dynamical diffraction conditions [10], such as the incoming and outgoing conditions, sample thickness. It has already been demonstrated by Pennycook in the 2 dimensional (axial channeling) ALCHEMI technique that the greater sensitivity of axial channeling allows the electron-channeling analysis to be extended to situations in which lower X-ray intensities are generated [11,12]. The sensitivity of axial channeling conditions might also bring new opportunities when combined with EMCD technique. For example, the site-specific EMCD technique [13,14], which is powerful to resolve complex magnetic structure and obtain quantitative magnetic parameters assisted with dynamical diffraction effects, has a critical demand for crystallographic structure. Proper diffraction conditions with enhancement of EMCD signals from different crystallographic sites are needed. Thus, its rich – rather than complex – dynamical diffraction effects will be an advantage here for site-specific EMCD. Secondly, the zone axial geometry has higher symmetry because of the symmetry of diffraction pattern. The effect of asymmetry of dynamical diffraction effects on signals acquisition and quantitative magnetic measurement has already been reported [10,15–20]. The ZA geometry can reduce or even eliminate the asymmetry in essence. Lastly, the two-beam or three-beam case of EMCD technique do not fit with many of other advanced and popular TEM characterization techniques which are always performed under the ZA conditions. Developing the EMCD technique under ZA geometry will provide the possibility of achieving collaborated characterization for information of local atomic structure, electronic structure and also magnetic properties. Especially, for ultrathin materials, such as the 2 dimensional materials, the sample stays at the ZA condition with high stability and it is difficult to be tilted to two-beam or three-beam case. Thus, the ZA geometry should be the only choice.

A requirement of a specific diffraction geometry is strongly desired to be overcome during the development of EMCD technique, since it puts up such restriction on single crystal that limits the wide application. By acquiring the signals under the random alignment of incoming beam and detector orientation with respect to the lattice axis of the polycrystalline iron, Muto et al. [21] introduce a statistical technique with new experimental geometry for the EMCD measurement of polycrystalline, but along with low SNR and complex signal processing. Although electron vortex beam provides us a new way to achieve magnetic measurement with no restriction on diffraction geometry, substantial progress in experiments have not been made according to the recent reports [22–25]. Therefore, performing the EMCD technique under different diffraction geometries, such as the quite general ZA case, is still applicable and meaningful.

Here, with the help of complex calculations of dynamical diffraction effects, the distribution of EMCD signals under the zone axial diffraction geometry is given in the diffraction plane, taking the example of yttrium iron garnet ($Y_3Fe_5O_{12}$, YIG), a typical magnetic insulator oxide with complex crystallographic and magnetic structure that is also used in the zone axial ALCHEMI technique [26]. The experimental diffraction geometry is designed and optimized for the acquisition of EMCD signals. At last, we demonstrate the measurement of EMCD signals under the ZA case, which are well consistent with simulations. The influence of dynamical factors on EMCD signals are discussed both in theory and experiments, including the incident conditions, detector positions and sample thickness. Especially, making full use of the very sensitivity of dynamical diffraction conditions, we experimentally obtain the separate EMCD signals of element Fe in YIG at each crystallographic site with the help of detailed analysis of dynamical diffraction effects.

2. Theory

The theoretical frame of EMCD and dynamical diffraction effects has already been quantitatively discussed in previous literature [13,20,27–32] and here we give a simple introduction. The EMCD experiment consists of taking two electron energy-loss spectra (EELS) at particular positions in the diffraction plane and the EMCD signal is the difference between these two EELS spectra. The double differential scattering cross-section (DDSCS) at opposite chiral positions of '+' and '−' on the diffraction plane could be written as (a simplified form that is suitable for cubic crystal structure, the detailed definition of variables in the following equation can be found in Refs. [13] and [20]):

$$\left(\frac{\partial^2\sigma}{\partial E\partial\Omega}\right)_{\pm} = \sum_u [\mu_+(E) + \mu_-(E) + \mu_0(E)]_u \cdot \frac{2}{3}\sum_{q,q'} \frac{q_x q'_x + q_y q'_y + q_z q'_z}{2q^2 q'^2} e^{i(q-q')\cdot u} \times \mathrm{Re}(A_{q,q'}) \pm \sum_u [\mu_+(E) - \mu_-(E)]_u \cdot \sum_{q,q'} \frac{q_x q'_y - q_y q'_x}{2q^2 q'^2} e^{i(q-q')\cdot u} \times \mathrm{Im}(A_{q,q'}) \quad (1)$$

By defining the terms of momentum-transfer and Bloch coefficients as the weighting factor (dynamical coefficients) of the intrinsic signal, the simplified formula is as follows [20]:

$$S_{\pm} = \frac{1}{2}\sum_u [\alpha_u \cdot (\mu_+ + \mu_- + \mu_0) \pm a_u(\mu_+ - \mu_-)] \quad (2)$$

u represents the coordinates of different atoms at different positions in a unit cell, α_u and a_u are the dynamical coefficients, S_+ and S_- are the EELS spectra acquiring from the conjugate positions in the diffraction plane respectively. $\mu_+(E) + \mu_-(E) + \mu_0(E)$ is the isotropic nonmagnetic signal and $\mu_+(E) - \mu_-(E)$ is the magnetic signal. The EMCD signal is the difference of the two spectra

$$EMCD = S_+ - S_- = \sum_u a_u(\mu_+ - \mu_-) \quad (3)$$

The relative intensity of EMCD signals is defined in Eq. (4) [14]. Thus, it is proportional to the dynamical coefficients, and we use a simple formula to represent the distribution of relative intensity of EMCD signals as shown in the following parts:

$$I = \frac{S_+ - S_-}{S_+ + S_-} = \frac{\sum_u a_u \cdot (\mu_+ - \mu_-)_u}{\sum_u \alpha_u \cdot (\mu_+ + \mu_- + \mu_0)_u}. \quad (4)$$

3. Crystallographic structure of YIG

To fully demonstrate the influence of dynamical diffraction effects on EMCD signals, the material system yttrium iron garnet ($Y_3Fe_5O_{12}$, YIG) with complex crystallographic and magnetic structure is chosen, which has already been used in the axial channeling ALCHEMI technique and its dynamical effects have been partly discussed [26]. More importantly, the EMCD signals for YIG under the planar channeling conditions have already been reported both in experiments and theory, and can be used here to make a comparison directly [14].

YIG ($Y_3Fe_5O_{12}$) has a ferrimagnetic garnet structure with a