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Site-specific chirality in magnetic transitions

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ABSTRACT

We report site-specific energy loss magnetic dichroism measurements of the technologically interesting Heusler alloy Ni_2MnSn. In addition, we confirm the theoretical prediction that under certain conditions, two different atoms on inequivalent lattice sites give dichroic signals with opposite signs. With this, it is possible to distinguish the magnetic moments of atomic columns that are merely 1.5 Å apart using a conventional transmission electron microscope without the need for aberration corrections.

1. Introduction

The discovery of magnetic circular dichroism in the transmission electron microscope (TEM) [1] provided an unexpected alternative to X-ray circular dichroism (XMCD) in the synchrotron. "Energy Loss Magnetic Chiral Dichroism" (EMCD), as it was called, has nanometer resolution [2] and—compared to XMCD in the synchrotron—easy access to additional information in one "beam line" via standard analytical techniques in the TEM [3]. The main difficulty of EMCD—the notoriously low signal—has been improved in the last years by using optimized scattering geometries [4]. EMCD has evolved into several techniques, now utilizing either energy filtering, spectroscopy, TEM, or scanning TEM conditions [5].

The basic mechanism underlying EMCD is the same as for XMCD: the excitation of an electronic transition with a change of the magnetic quantum number of an atom by ± 1 (chiral transition). In XMCD, this is achieved by using circularly polarized X-rays [6–9]; EMCD exploits differences in the double differential scattering cross section (DDSCS) of fast electrons for left- and right-handed chiral transitions [1,5].

In the experiment, a ferromagnetic specimen is illuminated with a superposition of plane electron waves under particular diffraction conditions, forcing chiral electronic transitions that obey the dipole selection rule for the magnetic quantum number $\Delta m = \pm 1$ or $\Delta L_z = \pm \hbar$. The particular condition of diffraction is realized by tuning the excitation error for the eigenfunctions of the probe electron in the crystal (the Bloch waves). It is determined by the demand that the two plane wave components with highest excitation strength exhibit a phase shift of $\pm \pi/2$ at the atomic sites of interest. In the standard 3*d* ferromagnets Fe, Co, and Ni, all atomic sites in a chosen depth of the specimen see the same phase shift and so contribute to the EMCD signal with the same sign. In this respect EMCD is similar to XMCD.

More generally, X-ray absorption spectroscopy is not site-specific. On the other hand, spectroscopic methods in the TEM have been used to obtain site-specific information on elemental concentrations and local densities of states [10,11], using channeling conditions in the crystal. The idea to combine site specificity with EMCD is not far fetched and has been considered theoretically [12,13]. The basic idea is that in a crystal with basis and, e.g., two different atomic species at non-equivalent crystallographic sites A and B, the two strongly excited plane wave components have different relative phase shifts. Under not very stringent conditions, one can see a phase shift close to pi/2 at site A and the opposite phase shift at site B. If these sites are occupied by different elements with ferromagnetic coupling, as it is the case, e.g., in Heusler alloys, an EMCD experiment will result in magnetic signals with opposite sign for the two elements, in contrast to XMCD. Therefore, the combination of channeling conditions and EMCD allows obtaining information on the position and the magnetic properties of the different elements inside the unit cell of complex crystals with a conventional electron microscope with a non-focussed beam. A site selective probe of

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magnetic moments [14] as described here bears promise for the study of magnetic order on the atomic scale, such as interfaces or dead layers, and can have impact on many fields, including nanomagnetics and spintronics.

In this paper we want to show in a demonstration experiment that a magnetic footprint of different atomic columns not more than 1.5 Å apart is readily accessible with a conventional TEM, and that the EMCD sign is indeed reversed for two inequivalent sites as expected from simulations. As a technologically interesting test material, a Ni_2MnSn Heusler alloy has been chosen for the model experiment. This complex ferromagnetic alloy is currently under investigation worldwide due to its interesting physical properties: shape memory and extraordinary magnetocaloric effects, as well as giant magnetoresistance will allow future applications in actuators, switching devices, magnetic cooling and hybrid systems [15–17].

2. Simulations

It is well-known that not only the incident beam, but also the outgoing beam must be considered if one endeavors to understand the origins of the EMCD signal [18–20]. As such, careful tuning of both beams—together with a suitable sample—can be exploited to achieve high site-selectivity, similar to the "energy-loss by channeled electrons" (ELCE) technique [10,11]. Obviously, numerical simulations are essential to find suitable parameters and understand the results.

To that end, we performed several calculations using a program for EMCD simulations [20] that uses the DDSCS in the form [5,18,20]:

$$\frac{\partial^2\sigma}{\partial E\partial\Omega} = \frac{4\gamma^2}{a_0^2}\frac{k_f}{k_i}\sum_{\mathbf{ghg'h'}}\frac{A_{\mathbf{gh}}A^*_{\mathbf{g'h'}}S(\mathbf{Q},\mathbf{Q}',E)}{(QQ')^2},$$

where γ is the relativistic factor, a_0 is the Bohr radius, k_i and k_f are the wave vectors of the probe electron before and after the inelastic scattering event, the A are Bloch wave factors, $\mathbf{Q}$ and $\mathbf{Q}'$ are the momentum transfers, and S is the mixed dynamic form factor (MDFF) [1,21,22], which was modeled in dipole approximation here.

We used, according to the experiment, a Ni_2MnSn lattice structure (compare Fig. 3) with lattice parameter $a=0.6051$ nm [23], a zone axis of [0 0 1], a systematic row condition including the (2 0 0) diffraction spots, an acceleration voltage of 300 kV, and a 5-beam incident/4-beam outgoing case [20]. As an approximation incoming plane waves have been used for simulations.

Fig. 1 shows which parts of the sample contribute in which way to the total EMCD signal for an ideal setup [20]. It can be seen that the Ni atoms are sitting at the border of the red region, indicating parts of the sample from which strong positive EMCD intensities originate. Likewise, the Mn atoms are sitting at the rim of the blue region, indicating strong negative EMCD intensities. This immediately explains the opposite signs of the EMCD signals on Mn and Ni atomic columns, respectively.

In addition, we have studied the thickness dependence of the EMCD signal to determine an optimal sample geometry. Fig. 2 shows the thickness dependence for Ni_2MnSn. It is clearly visible that the signal depends in a non-trivial way on the sample thickness. Also, it can be seen that for the specimen used in the experiment, theory predicts an EMCD signal of 16% for Ni and of -17% for Mn. However, the experimentally measured dichroic signal is expected to be somewhat smaller than these theoretical (maximum) values as a consequence of using a large circular spectrometer entrance aperture (SEA) which selects not only the intensity from positions in the diffraction plane that are ideal for EMCD measurements but also further experimental challenges like non-perfect crystal orientations, contamination, or beam instabilities.

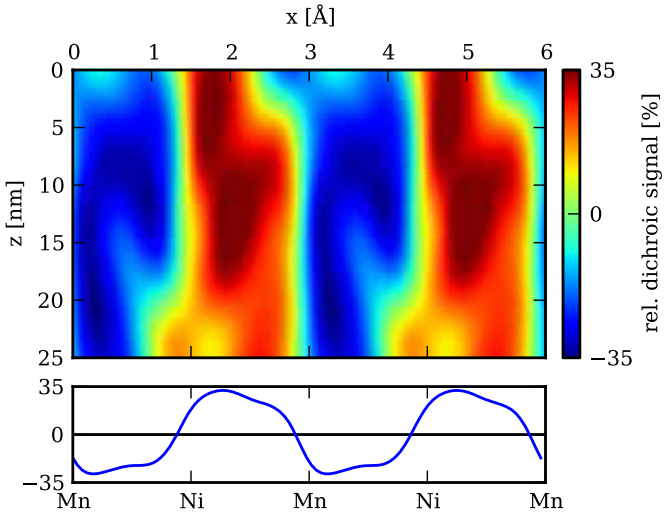

Fig. 1. The x–z map of the EMCD contributions of different regions of the sample (top) and z-integrated EMCD signal as function of position x in the unit cell (bottom). The image shows one (projected) unit cell horizontally, and the whole crystal of 25 nm vertically. The positions of the magnetic atoms are shown below the x-axis.

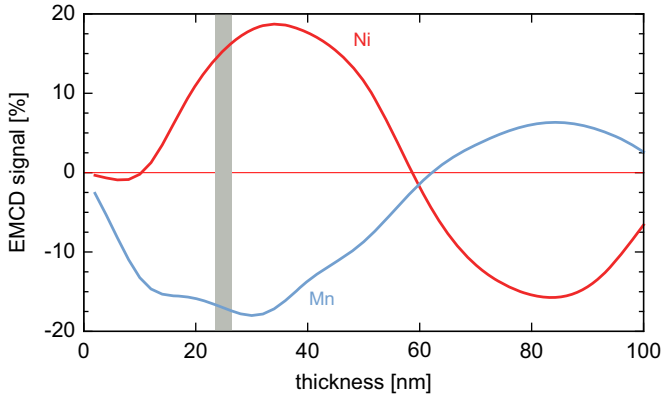

Fig. 2. Thickness map for the EMCD signal strength as function of the sample thickness. The thickness of the sample used in the experiments is indicated by the vertical grey bar.

3. Experiment

A 100 nm thick layer of the Heusler alloy has been grown by sputter deposition onto a MgO substrate and a cross section sample has been prepared by a classical grinding and ion milling process. The crystallinity of the sample in the interface region between the Ni_2MnSn layer and the MgO substrate can be seen in the HRTEM image shown in Fig. 3.

The EMCD experiments were performed in the scanning mode of a FEI Titan 80-300 TEM at room temperature and, therefore, in the austenite phase of the Ni_2MnSn sample [24,17]. The sample is magnetically saturated in the $\sim$2 T field of the objective lens at the sample position. The specimen was tilted into the three beam case (3BC) by minimizing the excitation error (compare Fig. 4(a)). The convergence angle was set to 3.25 mrad to ensure non-overlapping diffraction discs. The beam diameter on the specimen was about 1 nm. The axis of the 3BC was aligned perpendicular to the energy dispersive axis of the EEL spectrometer and the diffraction discs shifted out of the 2 mm SEA. Hereby, one half of the symmetry plane defined by the axis of the 3BC (see Fig. 4(b)) is selected and the differences of the chirality can be recorded simultaneously in a single EELS measurement [25,26].