Characterization of magnetic domain walls using electron magnetic chiral dichroism

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Abstract

Domain walls and spin states of permalloy were investigated by electron magnetic chiral dichroism (EMCD) technique in Lorentz imaging mode using a JEM-2100F transmission electron microscope. EMCD signals from both Fe and Ni L\(_{3,2}\) edges were detected from the Bloch lines but not from the adjacent main wall. The magnetic polarity orientation of the circular Bloch line is opposite to that of the cross Bloch line. The orientations of Fe and Ni spins are parallel rather than antiparallel, both at the cross Bloch line and circular Bloch line.

Keywords: electron magnetic chiral dichroism (EMCD), magnetic domain wall, permalloy, electron energy loss spectroscopy, Lorentz electron microscopy

1. Introduction

Spin-dependent transport of charge carriers has been extensively studied in tunneling magnetoresistance systems owing to their immense potential for magnetoelectric applications and a variety of interesting physical properties \([1–3]\). Thin ferromagnetic NiFe layers are usually used as electrodes in such systems. The magnetic domain walls inside these ferromagnetic layers are crucial for determining the tunneling spin polarizations and are particularly associated with the magnetic long-range order in a 3D Heisenberg system \([4]\). Undoubtedly, information on the wall types in magnetic thin films is important for various magnetic devices.

Efficient techniques of characterizing the magnetic properties of ultrathin electrode films with high spatial resolution are still lacking. Only a few direct studies have been reported on the spin states of individual domain walls in thin ferromagnetic layers, and there is still no consensus on how to reliably determine the type of an individual wall in a magnetic domain. Most methods used for this purpose, such as Bitter powder technique, magneto-optical Kerr effect, Lorentz microscopy, electron holography, magnetic force microscopy and x-ray topography, have insufficient selectivity or spatial resolution to directly relate the microstructure and magnetic character of individual domain walls \([5–7]\). In our previous work, the movement of the circular Bloch line was studied by \textit{in situ} Lorentz microscopy \([8,9]\). The magnetic contrast of Lorentz microscopy is based on the Lorentz force, which can only be used to identify the in-plane domains and Neel walls with magnetization orientation in the plane of a thin film. Conventional Lorentz electron microscopy mainly relies on the Foucault imaging mode and Fresnel-defocus imaging mode. There is no method of magnetically imaging a domain with magnetization out of the film plane, although such domains might be used for magnetic recording.

Circular dichroism is caused by the existence in the medium of some physical properties related to handedness and electron spin. The EMCD technique is based on electron energy loss spectroscopy (EELS) detection, which can provide chemical information at sub-nanometer scale, such as type, coordination and valence states of the constituent...
chemical elements. Lorentz microscopy can determine the orientation of a domain wall within a thin film, but not normal to the film plane. The latter orientation can be determined with EMCD because its detection depends on the orientation and strength of magnetization along the beam direction of a transmission electron microscope (TEM). The limitations of EMCD include incompatibility with automatic beam scanning due to the requirement of two-beam diffraction condition, as well as difficulty of operation, weak signals and dependence of the signals on the sample thickness. The magnetic field of the objective lens in a TEM would align the magnetic moments in the sample parallel to the optical axis (i.e. out of the sample’s plane). Therefore, an EMCD experiment has to be carried out in Lorentz mode, with the objective lens switched off.

The EMCD technique, based on electron energy loss near-edge structure (ELNES), similar to x-ray magnetic circular dichroism (XMCD), was proposed in 2003 [10] and experimentally developed in 2006 [11]. Momentum transfer in EMCD is considered to be equivalent to the polarization vector in XMCD, allowing the measurement of the magnetic circular dichroism without spin polarization of an electron beam. Chiral electronic transitions can be detected with a standard TEM equipped with an EELS system or an energy filter. Recently, Schattschneider and co-workers have reported a series of EMCD studies [12–15]. In particular, they explored the magnetic properties of thin films and individual nanoparticles by EMCD and dichroic map simulation [15]. Owing to its high spatial resolution, EMCD is becoming a key microscopic characterization method in spintronics and nanomagnetism [16,17].

In this work, we report an EMCD study of the cross Bloch line and circular Bloch line, in the case when the magnetization is directed out of the film plane. Based on the EMCD data, nanosized regions on the domain wall could be easily assigned to a cross Bloch line, a circular Bloch line or a Neel line.

2. Experimental details

A thin permalloy film was chosen as a typical ferromagnetic specimen. Permalloy has many advantages such as high magnetic permeability, low coercivity and magnetostriction and significantly anisotropic magnetoresistance. Permalloys typically have a face-centered cubic crystal structure with a lattice constant of approximately 0.355 nm for a nickel concentration of about 80%. A two-beam diffraction condition required for the EMCD experiment is relatively easy to achieve in such material. TEM samples were prepared by mechanical polishing and dimpling. An ion milling system (Gatan-691) with a liquid-nitrogen-cooled stage was used for thinning the samples. The in situ Lorentz microscopy examinations were carried out using a JEM-2100F TEM equipped with a field emission gun, with the objective lens switched off and the objective minilens on. A postcolumn Gatan imaging filter (GIF-Tridium) was used for EELS measurements. The energy resolution was ~0.70 eV, as evaluated using the full width at half maximum of the zero-loss peak. To avoid electron channeling effects, the selected grain was slightly tilted off the zone axis (by 2–4°). The convergence angle was ~0.7 mrad ($q \approx 0.04 \text{ Å}^{-1}$) and the collection angle was ~3 mrad ($q \approx 0.17 \text{ Å}^{-1}$).

3. Results and discussion

3.1. Model of cross tie

The structure of cross-tie walls was established by Hebert and Schattschneider in 1958 using Bitter pattern methods [10]. The Neel wall is spaced at regular intervals by short cross Bloch lines, denoted as ‘cross ties’, each terminating in a free end as shown in figure 1(a). Domain wall creeping of the circle-tie walls is connected with circular Bloch line movements and a variation of the main wall curvature [8,9]. Figure 1(b) shows an underfocal Lorentz microscopy image of a cross-tie wall. The dark line corresponds to a main domain wall—a 180° Neel-type wall. The two shorter white lines are
cross ties and therefore the intersecting points are cross Bloch lines, as marked by the arrows. The dark points between the cross ties are circular Bloch lines. Figure 1(c) presents another Lorentz image from the same specimen, showing a circular Bloch line with a stronger contrast.

To confirm that the dark and bright lines are magnetic domain walls, two sequential images were recorded with an underfocus of about $-25 \mu m$ and an overfocus of $+25 \mu m$. The transmitted electrons are deflected by the Lorentz force, the vector of which is tuned by the defocus values. Hence, ‘convergent’ or ‘divergent’ wall images were formed as shown in figure 2.

3.2. EMCD at cross Bloch wall and Neel wall

EELS examinations were performed to detect magnetic dichroism at several locations on the permalloy film. As in [11], EMCD signals are an order of magnitude weaker than XMCD signals. In our experiment, we used a condensed beam (spot size 5) in convergent-beam electron diffraction (CBED) mode. The angular convergence and the focal values of the incident beam were controlled by adjusting both the condenser lens and the prefield of the objective minilens in Lorentz mode. In two-beam CBED, an aperture selects only the central (000) beam and diffracted (111) beam $G$ in the back focal plane, as shown in the inset of figure 3.

A Thales circle was made up of the coherent transmission and diffraction beams and was drawn with a diameter $G$ to intersect the 0 and $G$ disks. The strongest dichroic signals can be detected at the top and bottom points on the Thales circle, which, respectively, correspond to $q^+$ and $q^-$ wave vectors. The EELS detector was positioned at the two moment transfer points. From the diffraction dynamic point of view, the imaginary part of the mixed dynamic form factor has maxima at the top and bottom positions where pseudovectors are also maximum [11].

Figures 3(a) and (b) show the EMCD energy spectra around the Fe L$_{2,3}$ and Ni L$_{2,3}$ edges measured from the cross Bloch line of a permalloy film. The existence of an EMCD difference signal confirmed that the Bloch line carries a magnetic flux parallel to the electron beam. The EMCD signals (normalized spin-up/down difference $\Delta > 0.06$) are strong enough to be visible without magnification. Excitations detected at the spin–orbit split 2p$_{3/2}$ and 2p$_{1/2}$ levels generated two peaks at $\sim 708$ and $\sim 721$ eV, respectively. This dichroic spectroscopy at Fe-L$_3$ and Fe-L$_2$ edges results from spin population imbalance around the Fe-3d Fermi surface. There are more spin-up electrons than spin-down electrons when detected at the $q^+$ position in momentum space. Combining the analysis of figures 1, 3(a) and (b), the orientation of the cross Bloch line was determined as parallel to the electron beam. The signal from electrons at spin-up states is higher than that from electron at spin-down states. In contrast, at the main domain wall, the magnetic flux is within the sample plane, which makes an orthogonal orientation relationship with the incident beam direction and therefore contributes
Figure 4. Typical EMCD spectra near the (a) Fe-L$_{2,3}$ and (b) Ni-L$_{2,3}$ edges recorded at a Neel wall, showing absence of EMCD difference signal.

Figure 5. Typical EMCD spectra near the (a) Fe-L$_{2,3}$ and (b) Ni-L$_{2,3}$ edges recorded at a circular Bloch line.

no dichroic signal to EMCD, as shown in figures 4(a) and (b).

3.3. EMCD at circular Bloch wall

The most remarkable phenomenon, revealed from the EMCD difference signal in figures 5(a) and (b), is that the spin orientation at the circular Bloch line is antiparallel to that at the cross Bloch line. The signal from electrons at spin-down states is stronger than that from electrons at spin-up states. However, the EMCD difference is weaker for the circular Bloch line than cross Bloch line ($\Delta_1 \approx 0.02$), which is discussed as follows. The magnetization orientation in each domain is determined by the competition of magnetostatic energy, magnetoelastic energy, demagnetizing field energy, exchange interaction, magnetocrystalline anisotropy, magnetic dipolar interaction and domain wall energy [5]. The circular Bloch line has a more complicated spatial distribution, which is similar to a three-dimensional spiral contour map, than the cross Bloch line. While the EMCD signal from the circular Bloch line is an average over the circle region, this is not the case for the cross Bloch line.

The spin-up signal is stronger than the spin-down signal at the Fe-L$_3$ edge at 708 eV. Similarly, the Ni-L$_3$ signal is also higher at the ‘+’ position (spin-up) than at the ‘−’ position (spin-down). Thus, a ferromagnetic order at both the cross Bloch line and circular Bloch line was deduced on the basis of the same spin orientation of Fe and Ni signals. This is because of metallic bonding between Fe and Ni atoms in permalloys, with a parallel spin orientation. Such Fe-Ni metallic bonding is composed of Fe 3d electrons with spin-up (spin-down) orientation and Ni 3d electrons with spin-up (spin-down) orientation.

4. Conclusions

We have combined convergent-beam electron diffraction and electron magnetic chiral dichroism in Lorentz mode to determine the nature of individual magnetic domain walls with a transmission electron microscope. This technique allows distinguishing the cross Bloch line, circular Bloch line and Neel wall. Its high spatial resolution enables the study of spin-dependent transport across a domain wall.
Acknowledgments

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