Development and Application of Magneto-Optical Microscope Using Polarization-Modulation Technique

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1. Introduction
Magnetic Imaging using MO method

- Magneto-optical (MO) microscopes have been used as one of the most significant techniques for an observation of magnetic domain structures in magnetic materials.

- Recently, this technique attracts great attention as a powerful tool for visualization of invisible phenomena: e.g.,
  - spin-accumulation in nonmagnetic semiconductors (1,2)
  - magnetic flux intrusion in superconductors (3)-(5).

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Magnetic Imaging of stray magnetic field using MO method

Stray field from sample magnetize the garnet film locally, and the distribution of field can be detected by means of MO method, as a polarization image.
Example of Magnetic imaging using MO method
Advantages of MO imaging to other imaging techniques

- MO microscopes have technical advantages:
  - a short measuring time
  - a simple instrumental setup compared with other imaging techniques, e.g., a magnetic force microscope (MFM) (6), a superconducting quantum interference device (SQUID) microscope (7) and a Hall-probe microscope (8).
Magnetic Imaging

<table>
<thead>
<tr>
<th>Method</th>
<th>Quantitative Magnetic measurement</th>
<th>Spatial Resolution</th>
<th>Dynamic measurement</th>
<th>Measuring time for one image</th>
<th>Special sample treatment</th>
</tr>
</thead>
<tbody>
<tr>
<td>Scanning SQUID microscope</td>
<td>○:10^{-7}T</td>
<td>&gt; 1 μm</td>
<td>no</td>
<td>&gt; 1 min</td>
<td>no</td>
</tr>
<tr>
<td>Scanning Tunneling Microscope (STM)</td>
<td>×</td>
<td>&lt; 1 nm</td>
<td>no</td>
<td>&gt; 1 min</td>
<td>Surface treatment (cleavage, etc.)</td>
</tr>
<tr>
<td>Magnetic Force Microscope (MFM)</td>
<td>△</td>
<td>&lt; 10 nm</td>
<td>no</td>
<td>&gt; 1 min</td>
<td>no</td>
</tr>
<tr>
<td>Bitter Method</td>
<td>×</td>
<td>&lt; 10 nm</td>
<td>no</td>
<td>~10 msec</td>
<td>Deposition of magnetic materials</td>
</tr>
<tr>
<td>Lorenz Microscope</td>
<td>○</td>
<td>&lt; 1 μm</td>
<td>yes</td>
<td>~10 msec</td>
<td>Thinner the sample for TEM measurement</td>
</tr>
<tr>
<td>Magneto-Optical Microscope</td>
<td>○:10^{-5}T</td>
<td>&lt; 1 μm</td>
<td>yes</td>
<td>~10 msec</td>
<td>no</td>
</tr>
</tbody>
</table>

MO microscope has advantages shown above. In addition, it is easy to develop it with low temperature, magnetic field, etc, since the MO microscope is a simple technique based on optical microscope.
Conventional MO microscope

Slightly off (~4 deg) from cross-polarizer configuration
MO observation using cross-polarizer technique

- Light intensity is symmetrical for plus and minus magnetic field direction, so that no magnetic contrast can be observed.
- Angle between two polarizers should be slightly off (~4°) from 90 ° in order to get a contrast.
Problems in MO imaging

- Image is dark. Quantitative measurement of MO values, Faraday effect, Kerr effect in inhomogeneous samples is difficult.

- MO microscope using polarization modulation technique.

- Zigzag Domain structure in MO indicator film deteriorates images.

- Bi:YIG prepared by metal-organic decomposition method.
2. MO microscope using polarization modulation method
Polarization modulation technique using photoelastic modulator (PEM)

- Magneto-optical spectra have been measured using PEM which modulates retardation of light at $\rho$ rad/s to produce LP, LCP and RCP sequentially.
- Polarization rotation is given by detecting $2\rho$ component and ellipticity is given by $\rho$ component.
Retardation modulation using PEM

- P and A are linear polarizers, M photoelastic modulator (PEM), D a detector.
- PEM consists of an isotropic transparent material (quartz, CaF$_2$ etc.) and a piezoelectric vibrator made of quartz.
- If PEM is fed with HF electric field with angular frequency of $p$ [rad/s], standing wave of the acoustic sound which generates in the transparent material uniaxial anisotropy oscillating with angular frequency $p$ [rad/s], which in turn leads to appearance of $\Delta n$.
- Optical retardation $\delta = \frac{\Delta n l}{\lambda}$ is modulated with an angular frequency of $p$ [rad/s], therefore $\delta = \delta_0 \sin pt$. 
Schematic explanation of retardation modulation

- Fig. (a) shows time-variation of optical retardation $\delta$. If the amplitude $\delta_0$ takes a value $\pi/2$, positive and negative peaks of $\delta$ correspond to RCP (right circularly polarized light) and LCP (left circularly polarized light), respectively.
- If the sample shows neither rotation nor circular dichroism, the lotus of the detected electric field vector changes as LP-RCP-LP-LCP-LP, as shown in Fig. (b). The x-component does not change as shown in Fig. (c).
- If the sample shows rotation, the lotus varies as shown in Fig. (d) and the x-component oscillates with angular frequency of $2\pi$ as illustrated in Fig. (e).
- If circular dichroism exists vector length of RCP and LCP becomes different as shown in Fig. (f), leading to oscillation of x-component with angular frequency of $\omega$ [rad/s].
MO Spectrometer layout
Magneto-optical spectrometer

- Xe lamp
- Double monochromator
- Optical system
- Electromagnet
- Sample
- Power supply for electromagnet
- Preampifier
- Power source for Xe lamp
- Wavelength scan driver
- Lockin amplifier
- Lockin amplifier 2
- PEM controller
How to apply modulation technique to MO microscopy

- Conventional PEM employs a modulation frequency as high as 50 KHz, which exceeds the frame rate of CCD cameras and cannot be directly applicable to MO microscopy.

- In the retardation modulation technique, rotation produces difference in x-component of linear polarization (LP) and circular polarization (CP), while circular dichroism (=ellipticity) produces difference in the x-component of right circular and left circular polarizations.
Modulation technique using image processing

- It is thus elucidated as follows:
- MO rotation image can be obtained by an image processing to take difference between LP and CP images.
- MO ellipticity image can be obtained by an image processing to take difference between RCP and LCP images.
Novel MO microscope with retardation modulation

- Microscope: Olympus BH-UMA
- **CCD camera:** Hamamatsu C4880 (Cooled)
- Analyzer (fixed): Glan-Thomson MG*B10
- Objective lens: NeoSPlanNIC 10 × 50
- Rotatable quarter waveplate: ACP-400-700 (acromatic waveplate)
- Polarizer (fixed): Glan-Thomson (MG*B10)
- Bandpass filter: Interference filter (450, 500, 550, 600, 650 nm, BW=10nm)
- Light source: Halogen-tungsten lamp 20W
Principle of image processing

\[ E_2 = \text{ASQPE}_1 \]

\[
= \frac{1}{2} \begin{pmatrix}
1 & 1 \\
\sin \theta_F - i \eta_F \cos \theta_F & \cos \theta_F + i \eta_F \sin \theta_F
\end{pmatrix} \begin{pmatrix}
1 + i \cos 2\varphi & \sin 2\varphi \\
\sin 2\varphi & 1 - i \cos 2\varphi
\end{pmatrix} \begin{pmatrix}
E_x \\
E_y
\end{pmatrix}
\]

\[
= \frac{1}{2} \left\{ \cos \theta_F + \sin \theta_F - \eta_F \left( \sin (2\varphi + \theta_F) - \cos (2\varphi + \theta_F) \right) + i \left\{ \cos (2\varphi + \theta_F) + \sin (2\varphi + \theta_F) + \eta_F \left( \sin \theta_F - \cos \theta_F \right) \right\} \right\} E_x
\]

\[ I(\varphi) = \left( \cos \theta_F + \sin \theta_F - \eta_F \left( \sin (2\varphi + \theta_F) - \cos (2\varphi + \theta_F) \right) \right)^2 
+ \left( \cos (2\varphi + \theta_F) + \sin (2\varphi + \theta_F) + \eta_F \left( \sin \theta_F - \cos \theta_F \right) \right)^2 |E_x|^2 / 4 \]

\[
\psi = 0^\circ \quad \text{LP} \\
45^\circ \quad \text{RCP} \\
-45^\circ \quad \text{LCP}
\]
MO imaging using rotatable quarter waveplate as polarization modulator

Faraday rotation

\[ \theta_F = \frac{1}{2} \sin^{-1} \left\{ \frac{2I_{LP} - (I_{LCP} + I_{RCP})}{(1 - \eta_F^2)|E_x|^2} \right\} \]

\[ \theta_F \approx \frac{1}{2} \left\{ \frac{2I_{LP} - (I_{RCP} + I_{LCP})}{(1 - \eta_F^2)(I_{RCP} + I_{LCP})} \right\} \]

Faraday ellipticity

\[ \eta_F = \frac{1}{2} \left( \frac{I_{LCP} - I_{RCP}}{|E_x|^2} \right) \]

\[ \eta_F \approx \frac{1}{2} \left( \frac{I_{LCP} - I_{RCP}}{I_{LCP} + I_{RCP}} \right) \]

Evaluation of Faraday rotation

$$
\theta_F = \frac{1}{2} \sin^{-1} \left\{ \frac{2I(0) - \{I(\pi/4) + I(-\pi/4)\}}{(1 - \eta_F^2)|E_x|^2} \right\}
$$

$$
\theta_F \approx \frac{1}{2} \left\{ \frac{2I(0) - [I(\pi/4) + I(-\pi/4)]}{(1 - \eta_F^2)[I(\pi/4) + I(-\pi/4)]} \right\}
$$

$$
\eta_F = -\frac{1}{2} \left\{ I(\pi/4) - I(-\pi/4) \right\} |E_x|^2
$$

$$
\eta_F \approx -\frac{1}{2} \left\{ \frac{I(\pi/4) - I(-\pi/4)}{I(\pi/4) + I(-\pi/4)} \right\}
$$

CCD image (a) LP, (b) RCP, (c) LCP, Processed image (d) rotation (e) ellipticity
Sample

Y$_2$BiFe$_4$GaO$_{12}$ Film/glass sub. prepared by MOD method

Rectangular Dots array
Size $50\mu m \times 50\mu m$
Thickness 200nm

An optical microscope image

Transmittances of glass substrate and garnet dot are quite different.
It is hard to obtain quantitative magnetic contrast by conventional MO imaging technique.
Image of Faraday Rotation

Magnetic field reversal.

Image of Faraday ellipticity

Reversal of magnetic contrast corresponding to magnetization reversal

$\lambda = 500 \text{ nm}$
Quantitative evaluation.

Rotation angle is obtained quantitatively.

\[ \lambda = 500 \text{ nm} \]

Magnetization reversal

Rotation angle is obtained quantitatively.
Magnetic field dependences of patterned garnet film measured with wavelength of 500 nm. Clear hysteresis loop was observed at garnet dot although no signal was obtained at glass substrate. Hysteresis data can be obtained for each pixels.
Averaging and smoothing

- 1 shot
  \( \sigma = 0.920^\circ \)

- 10 times accumulation
  \( \sigma = 0.148^\circ \)

- 10 times accumulation + smoothing
  \( \sigma = 0.048^\circ \)

\( \sigma \): standard deviation

\( \sigma \): standard deviation
**Faraday rotation and Faraday ellipticity spectra of patterned garnet film using MO microscope**

**Faraday rotation**

**Faraday ellipticity**

**Dots** show data measured by MO microscope using interference filter (450, 500, 550, 600, 650 nm) with band width of 10nm. **Solid lines** show spectra measured by MO spectrometer for garnet film without patterning.
The merits of the method

- **Merits**
  - Simultaneous measurement of rotation and ellipticity in one cycle of measurement
  - Quantitative evaluation of rotation and ellipticity is possible (standard sample is not necessary)
  - Faraday image can be clearly displayed even in the sample with inhomogeneous transmission
  - Magnetic hysteresis loops at any pixel point can be displayed, once MO images are acquired for a sequence of magnetic field swinging between negative and positive magnetic saturation.

- **Demerit**
  - This method takes a few tens of second to get one MO image.
MO imaging using LCM as polarization modulator

Liquid crystal: ZLI-4792
Substrate: ITO coated glass

MO microscope

CCD camera
- Hamamatsu C9300 201
- Number of Pixels $640 \times 480$
- Data transfer 150 frame/s

Computer
- CPU XEON 3.2GHz
- RAM 2GB

Interface
- AD-DA, GPIB, etc.

Software Development
- Visual Basic 6.0,
- Image capture SDK

Tishibashi et al., JAP, 100, 093903 (2006).
Sequence of MO Measurement

Voltage for LCM

Measurement

Data transfer

Integration

Calculation

Display

Delay time 200ms

RCP

V1

V2

V3

LP

LCP
Real time observation

0s 1s 2s 3s

4s 5s 6s 7s

8s 9s 10s 11s

Pattern size 50μm square
Sample Y$_2$BiFe$_4$GaO$_{12}$
3. MO indicator film
Requirements for MO indicator

- Large Faraday effect
  For visualize magnetic field
- Thin film with a thickness of ~ 1μm
  To detect magnetic field near sample before its distribution smears out.
- In-plane magnetization without magnetic domain
  For high resolution magnetic image
Problem with Domain Structure

If we use LPE garnet as an MO indicator, Zigzag-shaped magnetic domain appears in garnet film magnetized in-plane, which makes it difficult to observe a signal from a sample, especially in the case that a signal is small.
MOD process

- Cleaning of Substrates
- Spin Coating
- Drying
- Pre annealing
- Crystallization

Repeat

• MOD solutions (by Kojundo chemical lab.)
  made from carboxylic acids ~3 mol%

• Chemical compositions
  YBiFeO Y:Bi:Fe 2:1:5

• Substrate
  Gd₃Ga₅O₁₂ (111)

- Step 1: 500 rpm 5 sec
- Step 2: 4000 rpm 30 sec

- 150℃ 5〜60 min
- 450℃ 10 min
- 550℃〜900℃ 1 h
MO indicator film

$Y_{2-x}Bi_xFe_5O_{12}$/GGG(111)

Thickness 400 nm)

Magnetic field dependence of Faraday rotation
Problems are overcome with MOD indicator

MO indicator films without visible magnetic domain structure prepared by MOD (metal-organic decomposition) is suited for observation of small signal from the sample.
4. Magnetic imaging
   (1) Superconducting film
The magnetic flux intruding into the superconductor is transferred to the indicator film. The perpendicular component of the magnetization is observed by Faraday effect.

**Sample setups**

- **Garnet film with Pt mirror**
- **Gd₃Ga₅O₁₂ substrate**
- **MOD Bi:YIG, 400nm in thickness**
- **Pt**
- **MgB₂ pattern**
  - Prepared by MBE
  - Patterned by photolithography
  - Thickness: 100nm
  - $T_c \sim 30K$

**Al₂O₃ substrate**

**Magneto-optical image**
Patterned MgB$_2$ film

Grown by NTT research lab.

Circle pattern
Diameter: 0.5mm

Square pattern
Size: 100$\mu$m $\times$ 100$\mu$m

Optical images
The image from the indicator side prevents direct optical image of circular dot due to Pt-mirror.

Optical image (×5) of MgB$_2$ pattern(0.5mm φ)

No direct optical image of MgB$_2$ pattern is observed due to Pt mirror.

Only the magnetic fluxes can be visualized.
The image from the indicator side prevents direct optical image of square dots due to Pt-mirror.

Optical image ($\times 10$) of MgB$_2$ square dots (100$\mu$m $\times$ 100$\mu$m)

Optical image ($\times 10$) after stacking with the indicator.
MO images of 500μm circle

24 Oe

46 Oe

123 Oe

735 Oe

368 Oe

0.1 Oe

T = 3.9 K
MO images showing intrusion of magnetic fluxes into an MgB$_2$ circular dot at T=3.9K.
MO images of 100μm square

T=3.9K
Quantitative magnetic image can be obtained from MO image by using linear relation $\theta_F - B$ for the MO indicator film. Therefore, contrast in the image directly shows a magnetic field, $B$. 

Magnetic image of remanent state after application of Magnetic field of 735 Oe.
MO image of MgB₂

![Image of MgB₂]

**FIG. 4.** Temperature dependence of resistivity of the C-doped and ultrapure MgB₂ films plotted along with the pure MgB₂ film made by PLD (Refs. 5 and 16).


PLD-grown MgB₂

Oslo University
How to obtain current distribution from MO images

- **Ampére’s law**
  \[ \mu_0 J = \Delta \times B \]

- **Biot-Savart’s law**
  \[ B_z = \frac{\mu_0}{4\pi} \int \frac{(y - y')(J_x - (x - x')J_y)}{|r - r'|^3} dx' dy' \]

1) One uses models for current distribution and compare the calculated B with the measured one.

2) One directly inverts by numerical method.

It needs all B component, Bx, By, Bz, while MO images measures only Bz.
Inversion of Biot-Savart’s law
using convolution theorem

\[ B_z = \mu_0 H_{ex} + \mu_0 \int_V K_g(r,r')g(x,y)d^3r' \] ... (1)

\( g \): local magnetization
\( K_g \): green function
\( z \) component of magnetic dipole

Using convolution theorem
Eq.(1) can be transformed into

\[ \tilde{B}_z(k) = \mu_0 \tilde{K}_g(k)\tilde{g}(k) \]

\( x \) and \( y \) component of \( J \) are obtained as

\[ \tilde{j}_x = -i \frac{\tilde{B}_z}{\tilde{K}_x} \quad \tilde{j}_x = -\tilde{j}_x \frac{k_x}{k_y} \]

\[ \tilde{K}_x = \mu_0 \frac{e^{-kh}}{k} \sinh \left( \frac{kd}{2} \right) \left[ \frac{k_y}{k} + \frac{k_x^2}{k^2} \right] \]
Density of lines corresponds to current density. Color indicates local moment obtained in a calculation. 

Current density $\sim 6 \times 10^7 \text{A/cm}^2$
Nb pattern prepared on Bi:YIG

- Substrate $\text{Gd}_3\text{Ga}_5\text{O}_{12}(111)$
- MO indicator film $\text{Y}_2\text{BiFe}_5\text{O}_{12}$ (400nm) by MOD method
- Superconductor Nb (150nm) by sputtering method
- Mirror Au
- Pattern size of anti-dots 7, 10, 15$\mu$m

Optical image
MO images of 10mm anti-dots

MO images of Nb 10μm × 10μm anti-dots pattern with applying magnetic field. The sample was zero-field cooled down to 3.5K.
High resolution MO image
4. Magnetic imaging
   (2) Magnetic structures
Y-shaped patterns buried in Si

Cross sectional SEM image

SiO₂

Ni₈₀Fe₂₀

Si

Linearly aligned

Honeycomb aligned

0.9 μm

4 μm
MO Observation of Y-shaped patterns

SEM image

MFM image

MO images

Si sub

NiFe

4 μm

NA=0.6

05.07.27

5 μm

NA=0.85

05.12.05
Use of MO indicator for observation of in-plane magnetization
Conclusions

- Quantitative magnetic imaging by the MO imaging technique using the polarization modulation technique combined with MO indicator films was developed.
- This technique allows us quantitative and nondestructive measurements for magnetic stray field as well as current distribution.
- Evaluations of stray field, current distribution were demonstrated for the superconducting MgB$_2$ patterned sample.
Acknowledgement

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