Kerr effect microscope combined with a pulse magnet to observe high-entropy alloys fabricated using combinatorial technology

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ABSTRACT
We developed a module-type two-dimensional (2D) polar Kerr effect microscope using a pulse magnet with a pulse width of 13 ms and a maximum magnetic field of 7 kOe to accelerate the development of new soft magnetic materials. We also developed an algorithm and method to extract still images from a measurement movie file when the pulse magnetic field is applied. To evaluate the performance of this instrument, we measured high-entropy alloys based on FeCoMn samples, which were screened and selected using material informatics. Samples obtained through depositing Pt, Zn, Cu, and Ru on high-entropy FeCoMn alloys exhibited a brightness distribution proportional to the Kerr rotation angle in the applied magnetic field. The developed instrument enabled the high-speed magnetic field mapping of composition spread thin films and is expected to accelerate new material development.

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1. Introduction

The development of power semiconductor devices such as SiC and GaN has been progressing rapidly [1–4] for further energy saving and electrification. The development of soft magnetic materials exhibiting high frequency and low heat resistance to operate as power device systems is also important [5–7]. To accelerate the development of materials with multiple properties, the screening of new materials is performed at a high speed using material informatics (MI) based on information technology [8–11]. Next steps involving the rapid fabrication of materials screened using MIs and their measurements and evaluations are required. For example, Hitosugi et al [12] developed a robotics-based
integrated system in combination with Bayesian optimization for fabricating and evaluating materials to accelerate material development.

We have been seeking new material candidates based on composition-spread thin films (combinatorial samples) with a size of 0.5–2 in using the thin-film combinatorial method; the feature of this method is that the sample position corresponds to the composition [13,14]. Various two-dimensional (2D) mappings are useful for revealing the multiple properties of combinatorial samples. Constructing such an evaluation system requires standardizing protocols and interfaces such as an approach to samples and data output and miniaturizing and modularizing each device and connecting them.

To evaluate magnetic properties, some measuring instruments such as vibrating sample magnetometer (VSM) and the Kerr effect have been developed. The Kerr effect method does not require a mechanism to vibrate the sample as required in VSM, so the instrument configuration is compact. Commercially available Kerr effect instruments use a constant-current magnet to perform the magnetic domain imaging of ferromagnetic materials at a few micrometers and the hysteresis measurement of the Kerr signal and magnetic field strength at selected points. Some highly sensitive measurement methods for very thin films have been developed [15–17] recently. A common bottleneck in miniaturizing the Kerr effect instruments is that the magnets applying the magnetic field occupy most of the instrument space. Therefore, by changing this magnet to a pulse magnet, the size of the instrument could be significantly reduced and the degree of freedom in arranging the instrument could be increased. Pulse magnets have mainly been developed to obtain high magnetic fields (more than 10 T) that cannot be achieved using constant-current magnetic systems [18]. However, when using a weak magnetic field, the magnet capacity and power supply are smaller and more compact than normal usage.

In this study, by combining the Kerr effect method and a pulse magnet (applied maximum magnetic field of more than 5 kOe), we developed a small module-type instrument with 2D mapping for centimeter-order composition-spread samples (diameter of approximately 10 mm). The developed instrument was approximately 1/5 the size of commercially available instruments. We used soft magnetic materials based on high-entropy alloys as an evaluation sample. The sample comprising a high-entropy alloy based on FeMnCo with the addition of a fourth element (M), such as Pt, Cu, Zn, and Ru, was selected using MI [19–22] and prepared using thin-film combinatorial technology.

2. Developed instrument

The Kerr effect refers to the polarization of light when linearly polarized light reaches the surface of a magnetic material, and it is used as a measurement method for determining the material magnetization state. The developed instrument comprised a light irradiation component to irradiate polarized light to the sample, reflection component to reflect polarized light from the sample, pulse magnet component to

![Figure 1](image_url). Schematic and photographs of the developed instrument. The instrument is based on a stereomicroscope with a polarizing optical system, pulse magnet, and camera. The instrument is downsized, and the region indicated by the dotted line in (a) is within 30 cm (width) × 50 cm (height). The arrangement of this coil and optical system enables polar Kerr measurements.
apply a magnetic field, power supply for the coil, and measurement control and data processing component. Figure 1 shows a schematic (Figure 1(a)) and photographs (Figure 1b) of the developed instrument. The characteristics of each component are shown in Table 1. The instrument is based on a stereomicroscope with a polarizing optical system, pulse magnet, and camera. The measurement sample can be accessed from the top surface. The developed instrument was downsized, and the region indicated by the dotted line in Figure 1(a) was within 30 cm (width) × 50 cm (height). The arrangement of this magnet and optical system enabled polar Kerr measurements. The maximum pulsed magnetic field was approximately 7 kOe with a pulse width of approximately 13 ms. The sample size was approximately φ10 mm. The horizontal resolution derived from the measurement field of view and the number of pixels of the camera was 20 μm. The size of the instrument is approximately five times smaller than that of commercially available instruments (e.g., the size of NEOARK BH804P polar Kerr measurement unit was 180 cm (width) × 200 cm (height)).

2.1. Optical system

The optical system consisted of an incident optical system and reflective optical system, where light passes through a biconvex lens (focal length 50 mm) from a light-emitting diode light source (peak wavelength 625 nm and bandwidth 15 nm) through multimode fiber and a film-type polarizing plate (extinction ratio 1,000,000:1 at 623 nm). Light is directed to the sample through a beam splitter and coil hole and then reflected off the sample surface. At this time, the polarization angle changes owing to the magnetization characteristics of the sample. The reflected light passes through the film-type polarizer and then introduced to the camera through the stereomicroscope, which has a three-barrel type optical path (Greenough type). The stereomicroscope was tilted at 10° with respect to the coaxial incident illumination. The camera was a complementary MOS (CMOS)-type monochrome camera with 4 million pixels, maximum resolution of 720 × 540, and maximum frame rate of 100 fps. Considering the field of view of the device and the number of pixels of the camera, the horizontal resolution was approximately 20 μm.

2.2. Pulse magnet generation system

An air-core coil (solenoid) without a magnetic core was used because its core provides an optical path for incident and reflected light. Therefore, the measurement area was determined using the size of the center hole of the coil. The coil with a core diameter of 15 mm was fabricated using a wire diameter of φ1.4 in and 120 turns (DC resistance value 0.183 Ω, inductance 0.438 mH, and impedance 0.248 Ω at 35 Hz). The image of the coil is shown in Figure 1(c). The maximum magnetic field was approximately 7 kOe at a discharge voltage of approximately 100 V. The power supply had a 50-mF condenser (10 mF × 5) with a breakdown voltage of 250 V. Additionally, the interface between the computer and power supply was installed to control discharging and charging from the computer. Figure 2(a) shows the measurement results of the magnetic field using a pulse magnetic flux measuring instrument (Nihon Denji Sokki co., Japan, Pulse Tesla meter GP-300) as a function of the charging voltage. The magnetic field was nearly constant at the sample position. The pulsed magnetic field was linearly proportional to the applied voltage. Figure 2(b) shows the time distribution of the magnetic field at discharge at a charging voltage of 100 V and current simulation distribution of the LCR series discharge circuit. The measured pulse half width was approximately 13 ms, which agreed well with the

Table 1. Characteristics of each component of the developed instrument.

| Parts          | Specification                                                                 |
|---------------|-------------------------------------------------------------------------------|
| Camera        | USB3 Vision Camera (0.4 Mega pixel, Monochrome, 8bit, CMOS),                  |
|               | Maximum resolution 720 × 540, Frame rate 100 fps (720 × 540),                |
| Microscope    | 3 Lens barrel type stereomicroscope,                                        |
|               | Greenough optics, Objective lens magnification 0.67–4.5                     |
| LED Source    | 625 nm (Bandwidth FWHM)15 nm, 13.2 mW (Min) LED, 1000 mA                    |
| Multimode fiber | Nanoparticle film type linear polarizer, 510–800 nm,                        |
| Collimated lens | Polarization independent type, backside AR coating, 400–700 nm             |
| Linear Polarizer | Size of hollow core φ15, Wire diameter φ1.4 (120 turn),                 |
| Half mirror   | Polarization independent type, backside AR coating, 400–700 nm             |
| Coil          | 50 mF condenser (10 mF × 5), Breakdown voltage 250 V                        |

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LCR simulation. Because the pulse half width was approximately 13 ms, the camera frame rate of 30–60 fps (approximately 10–30 ms) was used.

### 2.3. Measurement flow and data processing

The measurement flow and data processing are described as follows. The program was prepared using Python [23] and its modules.

Measurement flow (power supply and camera controlled using the computer):

1. Set the charging voltage of the power supply and start charging
2. When charging is completed, start the video recording and current flow from the capacitor to the coil (discharge).
3. After the discharge is completed, stop the video recording.

Data processing flow (an algorithm for extracting the frame when the magnetic field is applied):

The algorithm of data processing is shown in Figure 3. The following processing is performed using the video image acquired by the measurement system to create an image difference between the frame when the magnetic field is applied and the background.

![Algorithm for the image data analysis](image_url)

**Figure 3.** Algorithm for the image data analysis.
(1) Divide the recorded video into each frame (still image) and calculate the average and standard deviation brightness for each frame.

(2) Select the frame of the maximum average brightness.

(3) Calculate the image difference between the maximum average brightness frame and the background frame. One is the frame when the magnetic field is applied, and the other is the background.

(4) Calculate the histogram averaging of the image difference to emphasize the contrast.

To achieve the above algorithm, the material that definitely brightens due to the applied magnetic field was placed together with the measurement sample as a reference in the sample holder. Using this method, it is possible to extract the frame by applying the magnetic field.

3. Experimental results and discussion

3.1. FeCoMn + M sample

A schematic diagram of the FeCoMn + M sample is shown in Figure 4(a). The base substrate was created using a 100-nm thermal oxide film on Si and alternately depositing 200 layers of FeCoMn and the fourth element (Pt, Ru, Zn, and Cu) at 0.25 nm. Figure 4(b) shows the result of the polar Kerr measurement using a commercially available instrument (NEOARK, Japan, BH804P polar Kerr measurement unit) with a constant-current magnetic system. The horizontal axis is the applied magnetic field, and the vertical axis is the Kerr rotating angle. Hysteresis loops could be observed, except for Ru. Generally, soft magnetic materials exhibit small coercive forces and reach saturated magnetic flux densities by applying low magnetic fields. However, owing to the large demagnetizing factor in the case of the polar Kerr arrangement, the slope of the spectrum was small. In this case, the saturation magnetic flux

![Figure 4. Schematic of FeCoMn + M (M = Pt, Zn, Cu, and Ru) samples (a). The base substrate was created using a 100-nm thermal oxide film deposited on Si and alternately depositing 200 layers of FeCoMn and the 4th element (Pt, Ru, Zn, and Cu) at 0.25 nm. The result of the polar Kerr measurement using a commercially available instrument (NEOARK, Japan, BH804P polar Kerr measurement unit) with a constant-current magnetic field (b). The horizontal axis is the applied magnetic field, and the vertical axis is the Kerr rotating angle.](image-url)
density (maximum Kerr rotation angle) cannot be observed unless a magnetic field of 3 kOe or more is applied. Figure 5 shows the measurement results of our developed instrument. The four types of sample were placed in the sample holder. The applied magnetic field in this experiment was 7 kOe. The frame rate was 30 fps. Figure 5(a) shows the average and standard deviation brightness of each frame. The average and standard deviation brightness rapidly changed with the applied magnetic field. Figure 5(b) shows the image difference between before and after applying the magnetic field. It can be seen that Pt was bright and Ru was dark. Moreover, Zn and Cu were slightly bright. Because these samples were prepared using a soft magnetic material, Kerr rotation was only obtained when a magnetic field was applied and the brightness depended on the Kerr rotation angle. Figure 5(c) shows the plot of the brightness of Pt, Zn, Cu, and Ru with rotation angles. The rotation angles of Pt, Zn, Cu, and Ru were 0.18°, 0.1°, 0.06°, and 0°, respectively, at 7 kOe (Figure 4(b)). The obtained brightness was proportional to the rotation angles and agreed with the Kerr measurement shown in Figure 4(b).

3.2. FeCoMn + M composition-spread thin-film

A schematic illustration of the composition-spread thin-film sample is shown in Figure 6(a). To prepare the measurement sample, a 100-nm Si thermal oxide film was applied on a Si substrate and the FeCoMn sheet and Ru element were sputter-deposited at room temperature using a magnetron sputtering method. During the thin-film deposition, the thickness of a single composition gradient layer of FeCoMn and Ru was 0.5 nm. To form this layer, FeCoMn was first sputter-deposited on the substrate using a moving mask while controlling the volume. Next, Ru was similarly sputter-deposited using a moving mask to form one layer of the composition gradient film, and 200 such layers were deposited. The prepared sample is shown in Figure 6(b), and the unevenness was observed at approximately 13 wt% (weight percentage) composition ratio. Figure 7(a) shows a schematic of the measurement positions and corresponding wt% of the Ru components using the two measurement methods. Note that the dotted red rectangle was the measurement area of the developed instrument, and the red circle was the measurement point of the commercially available instrument with a beam size of 1 mm. FeCoMnRu50wt% and FeCoMnPt50wt% samples used in the measurement described in Section 3.1 were placed above and below the composition-spread sample as a reference. This was performed to identify the frame when a magnetic field is applied and correct
the Kerr rotation angle. The polar Kerr results using the commercially available instrument are shown in Figure 7(b). The Kerr rotation was observed at the Ru concentration of 0–13 wt% but not above 19 wt%. Figure 7(c) shows the measurement results using the developed instrument. The light and dark areas were clearly separated at the uneven region of the composition-spread sample. In Figure 7(b), the saturation magnetic field decreased at 13 wt%, which was the average value in both the regions due to the large beam diameter of 1 mm. From the 2D measurement using the developed instrument, the saturation magnetic field was almost the same under 13 wt% and disappeared above 13 wt%. In our 2D-X-ray diffraction measurement [22], phase separation was observed in this region. The 2D mapping measurement makes it easy to compare
it with other 2D characteristic mapping data such as 2D-X-ray, and it is useful in the evaluation of materials with various characteristics.

4. Conclusion

A module-type polar Kerr instrument with a 2D magnetic field (Kerr rotation angle) was developed. The entire instrument size was less than 1/5 the size of conventional instruments. The generated pulse magnetic field was 7 kOe with a pulse width of 13 ms. The horizontal resolution was approximately 20 μm per pixel. We created an algorithm and method to determine the change in the histogram value of the image to obtain a data image when the magnetic field was applied. In the samples obtained by depositing Pt, Zn, Cu, and Ru on high-entropy FeCoMn alloys, a brightness distribution proportional to the Kerr rotation angle in the applied magnetic field was obtained. The results obtained using the developed instrument agreed with those obtained using conventional instruments. The composition-spread thin-film sample created via the combinatorial method was also measured. Phase separation occurred at a Ru concentration of approximately 13–19 wt%, and the brightness distribution also coincided with the phase separation. The developed instrument enabled the high-speed magnetic field mapping of combinatorial samples and is expected to accelerate new material developments.

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Disclosure statement

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