

Magneto-optical Kerr-effect measurements under pulsed magnetic fields over 40 T using a compact sample fixture

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The magnetic field is one of the most fundamental control parameters in materials science. A pulsed magnetic-field apparatus can generate high magnetic fields that are inaccessible by conventional dc-field magnets. One important issue is that measurement techniques compatible with pulsed fields are rather limited due to short pulse duration and large electromagnetic or mechanical noise originating from field pulses. The magneto-optical Kerr effect (MOKE), the change in the state of light polarization upon reflection from magnetic materials, has the potential to become a powerful tool for investigation of magnetic properties of a wide range of materials including nontransparent materials or thin films in pulsed fields. Nevertheless, since the MOKE response is typically very small, MOKE measurements under pulsed fields are quite challenging. Here, we present a method to measure polar MOKE under high pulsed magnetic fields of 2-ms pulse width. The keys of this technique are a ferrule-based compact sample-fiber fixture and a phase-resolved numerical lock-in analysis, combined with the high-resolution optical apparatus based on an all-fiber loopless Sagnac interferometer. We succeeded in measuring MOKE signals from various ferromagnetic or ferrimagnetic samples above 40 T and down to 77 K, significantly extending the limits of previously reported pulse-field MOKE measurements. Our apparatus is simple enough to be compatible with larger-scale experiments in pulse-field facilities, thus becoming a promising tool to optically investigate material properties in pulsed fields.

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I. INTRODUCTION

The magnetic field is a fundamental control parameter in materials science. Under very high magnetic fields, the Zeeman energy experienced by an electron spin becomes comparable to the thermal energy. In such high fields, typically larger than tens of tesla, different phases of matter can be induced. Indeed, several field-induced phase transitions suggesting such unconventional mechanisms have been observed, e.g., the insulator-metal transition in VO₂ [1], the structural transition in solid oxygen [2], and exciton condensation in LaCoO₃ [3]. Therefore, strong magnetic fields open a frontier in materials research.

A pulsed magnetic field is a powerful tool to induce and investigate such high-field phenomena. While the world record for the highest static magnetic field is 45.5 T [4], the pulsed field in large facilities currently reaches even 1200 T [5]. In addition, in ordinary laboratories, a portable pulse-magnet system can now create fields larger than 40 T [6], which is

comparable to the world's highest static field. An important challenge in pulse-field experiments is that the typical duration of the pulsed field is on the order of milliseconds for a 100-T pulse and microseconds for a 1000-T pulse. Such short durations require measurements at frequencies on the order of megahertz and thus restrict the range of applicable techniques.

High measurement frequencies above megahertz can naturally be achieved by optical probes. When the light interacts with a material without the time-reversal symmetry, right- and left-circularly polarized light gain different phases. The cause of this difference in phase is called the magneto-optical (MO) effect. The MO effect can also be described as a rotation of the polarization of linearly polarized light. The MO effect seen via transmission is called the magneto-optical Faraday effect, and the effect observed via reflection is called the magneto-optical Kerr effect (MOKE). The polarization rotation angle due to the Faraday effect, the Faraday angle θ_F , can be as large as several radians for transparent samples with large thickness, and thus the Faraday effect has already been used in pulse-field experiments [5,7]. In contrast, pulse-field MOKE measurements are much more challenging, since the typical polarization rotation angle due to the Kerr effect, namely, the Kerr angle θ_K , is at most on the order of milliradians. Indeed, there are only a few examples of pulse-field MOKE studies, where measurements have only been performed up to 11 T and at room temperature [8,9].

On the other hand, MOKE experiments have led to various innovations in recent materials science. Fascinating examples include discoveries of chiral superconductivity

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accompanied by spontaneously broken time-reversal symmetry in the superconducting state of Sr_2RuO_4 [10], UPt_3 [11], and URu_2Si_2 [12]. The observation of the quantization of the Kerr angle in graphene [13], reminiscent of the quantum Hall effect, demonstrates the importance of the band topology in optical phenomena. The discovery of the first van der Waals ferromagnet CrI_3 was largely owing to MOKE measurements [14]. These studies also demonstrate a general advantage of MOKE over the Faraday effect, namely, that it is applicable to thin samples such as films and two-dimensional materials as well as to nontransparent materials such as conductive materials. Considering discoveries achieved by these previous MOKE studies and the wide applicability of MOKE, it is natural to expect that MOKE measurements in high pulsed fields, if realized, will yield further significant discoveries in high-field regimes.

To solve difficulties in realizing pulse-field MOKE due to the smallness of the MOKE response, we need a high-resolution measurement technique. Here, we came up with the idea to combine pulsed fields with the loopless (zero-loop-area) Sagnac interferometer [15], which has been used to detect subtle but nontrivial spontaneous magnetic signals in zero or small static fields [10–12, 16–19]. However, the very short timescale for measurements in pulsed fields urges us to develop another analysis method. In addition, we needed to develop a compact and stable setup that fits inside a small bore of a pulse-field magnet and is capable of overcoming electromagnetic and mechanical noises originating from the large current and Lorentz force generated by the pulse-magnet system.

In this article, we report a successful observation of polar MOKE up to 43 T, enabled by a compact sample fixture for optical measurements combined with a portable pulse-field system and a phase-resolved numerical lock-in analysis. To the best of our knowledge, our results record the highest magnetic field under which the Kerr effect has ever been observed. The sample fixture utilizes standard connector parts of optical fibers and provides a plug-and-play measurement free from complicated alignment processes of the incident light. Our setup, enabling quick magnetic measurements above 40 T in the laboratory, offers a route to explore magnetic properties of materials under extreme conditions.

II. FERRULE-BASED COMPACT SAMPLE FIXTURE

In optical measurements, including MOKE measurements, it is important to ensure a proper illumination of the sample and, further, to achieve optimal coupling of light reflected from the sample into the photodetector. In previous pulse-field MOKE studies, ordinary free-space optics were used and sample coupling was achieved by precise alignment of the optical path using lenses and mirrors [8,9]. Extensions of such free-space-based MOKE measurements to higher fields or to lower temperatures are not straightforward.

Instead of free-space optics, we intend to use a fiber-based loopless Sagnac interferometer for pulse-field MOKE measurements. In previous MOKE studies using the loopless Sagnac interferometer in dc fields, various setups, such as miniature tripod structures, have been used, so that the light reflected from the sample is coupled back to the

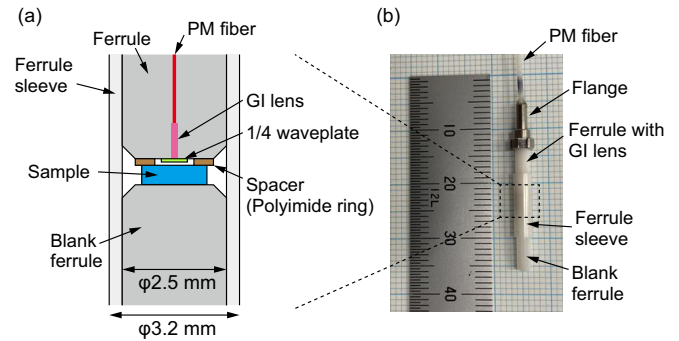


FIG. 1. (a) Schematic of the central part of the sample fixture composed of a ferrule-embedded GI lens focuser, 1/4 wave plate, ferrule sleeve, and polyimide spacer. The spacer thickness is chosen to be $50\ \mu\text{m}$, such that the sample surface is placed at the focal position of the GI lens simply by sandwiching the sample with two ferrules. The diameter of the ferrules is 2.5 mm and the outer diameter of the ferrule sleeve is 3.2 mm. (b) Photo of the whole fixture. The ferrule focuser and the blank ferrule are fixed using a ferrule sleeve.

polarization-maintaining (PM) fiber [17,19,20]. Nevertheless, such setups are generally too large to be placed in small bores (with diameters typically less than $\phi 10\ \text{mm}$) of pulse-field magnets [5,6]. In addition, under pulsed magnetic fields, a short and strong field pulse can cause vibrations in the setup, which would disturb the optical coupling. In addition, because a pulsed field generates Eddy currents in metallic parts, it is important to avoid highly conductive metals in the setup. Thus, it was necessary to invent a compact, stable, and non-metallic setup.

In this work, we propose a setup that is based on ceramic ferrules. Ferrules are small rods that are used in optical fiber connectors. They are made with high precision, making them suitable for optical coupling, although they are not costly. They are typically made with nonmagnetic zirconia ceramics, which is suitable for avoiding Eddy-current effects in pulsed magnetic fields. As shown in Fig. 1, we used a custom PM-fiber focuser made with a gradient-index (GI) lens embedded in a 2.5-mm diameter ceramic ferrule (Photonic Science Technology). This focuser has a focal length of $50\ \mu\text{m}$ and the diameter of the focused beam is around $6\ \mu\text{m}$. A 1/4 wave plate made of thin polyimide (NTT Advanced Technology Cooperation, AT-QWP-4A; $0.75 \times 0.5 \times 0.015\ \text{mm}^3$) is glued at the tip of the focuser. The optical axes of the wave plate are 45° tilted with respect to the slow/fast axes of the PM fiber, so that the linearly polarized lights from the focuser are converted to the free-space circular polarized lights, and vice versa. The sample is placed beneath the focuser, with a $50\text{-}\mu\text{m}$ -thick polyimide spacer. This spacer makes the focal point of the light match the sample surface. The sample is fixed inside a ferrule sleeve (Thorlabs, ADAF1), sandwiched by the ferrule focuser and by a blank ferrule (Thorlabs, CF128-10). One side of the sample should have good optical reflection, whereas the other side does not need to be optically flat, but should be parallel to the other side. If the surfaces are not perfectly parallel, it might be helpful to place a suitable cushion material between the sample and the blank ferrule.

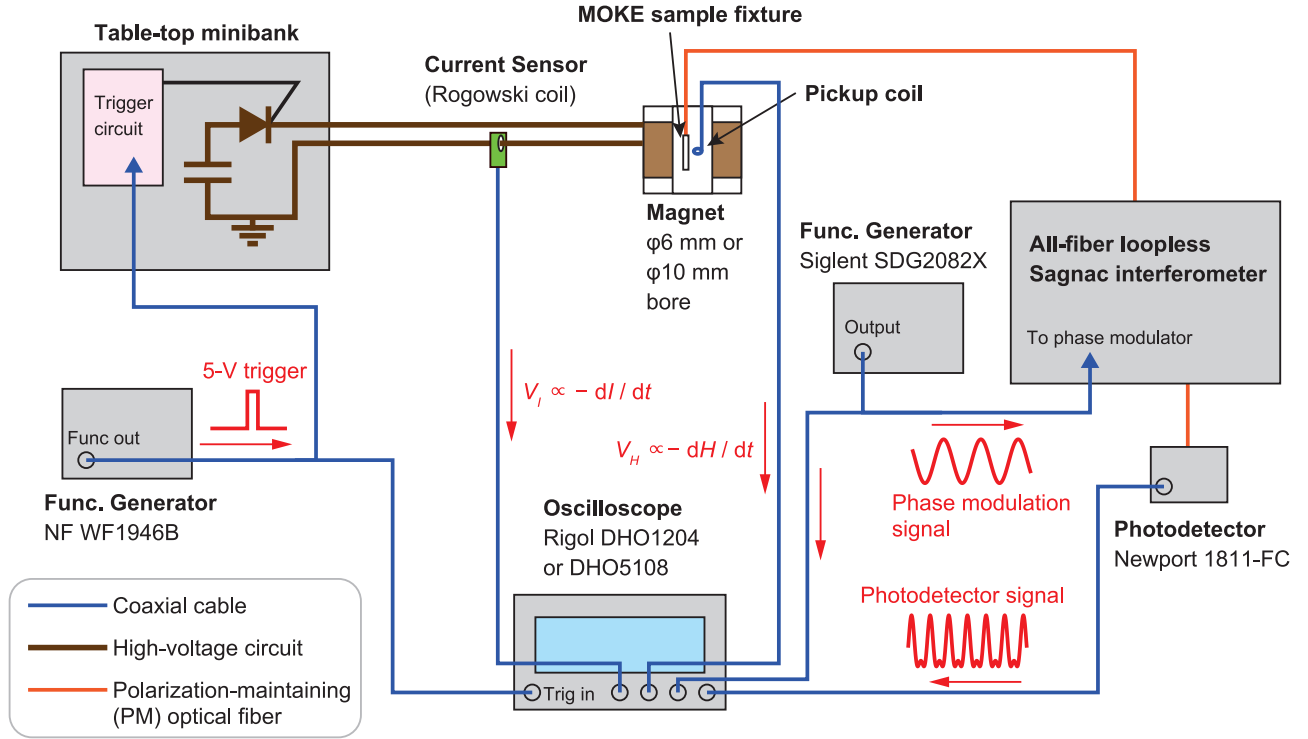


FIG. 2. Measurement setup for the magneto-optical Kerr effect under pulsed magnetic fields. Sets of voltage recorded by the oscilloscope are analyzed using the numerical lock-in analysis. When we use the 6-mm coil, the field pickup coil and the Kerr fixture cannot be simultaneously inserted to the bore. Thus, we first run calibration with the pickup coil in order to obtain the field-current relation. In subsequent MOKE measurements without the pickup coil, the field value is calculated from the magnitude of the current.

Figure 1(b) shows a photo of the entire ferrule-based fixture. The outer diameter of the ferrule sleeve is 3.2 mm. The maximal diameter of the setup is about 4.5 mm due to the metallic flange made of stainless steel. Since the electrical conductivity of stainless steel is relatively small, we do not notice any Eddy-current effects. The total length of the fixture is around 30 mm. Notice that it is possible to further minimize the fixture by using ferrules with 1.25-mm diameter and by avoiding the use of the metallic flange.

This fixture demonstrates very high and stable optical performance. We have found that more than 50% of the incident light is coupled back to the fiber without any precise adjustments, if we use samples with relatively high reflectivity. Thus, it only takes a few minutes to prepare for the experiments. For comparison, it typically takes more than 1 h to optimize the optical coupling with the previous setup [17]. This significant reduction of the preparation time is crucially important to achieve high-throughput experiments using pulse-field magnets, as well as other MOKE experiments using ordinary dc magnets. Moreover, the optical coupling is quite stable, and moderate mechanical shocks do not affect the intensity of the reflected light. We have also tested this fixture in low-temperature environments, such as through direct immersion in liquid nitrogen down to 77 K, or dilute gas helium in a commercial cryostat (Quantum Design, PPMS) down to 2 K. In both cases, the change in the coupling is found to be typically around 10%, as explained in Appendix A.

III. OPTIC AND ELECTRIC APPARATUS

For MOKE measurements, we used an all-fiber loopless Sagnac interferometer. The basic concept of our interferometer is the same as those reported in Refs. [10,15]. The light source is a low-coherence superluminescent diode (SLD) (Thorlabs, SLD1550P-A1 and SLD1550P-A40). The emitted light has a center wavelength of 1550 nm, corresponding to 0.8-eV photon energy. The overall light intensity of the interferometer was measured using an InGaAs photodetector (Newport, 1811-FC). The main difference with respect to Refs. [10,15] is that we replaced free-space optical parts with PM-fiber-based optical instruments, such as a variable wave plate with a polarizer and a 45° principal-axis splice (Phoenix, PWP-02-15-PM-2-1) and a phase modulator (iXblue Photonics, MPX-LN-0.1). This all-fiber setup has the advantage of being easy to build and highly stable against mechanical vibrations, etc. In addition, fiber-based phase modulators operate in a much wider frequency range than free-space phase modulators.

Figure 2 illustrates our experimental setup. The pulsed magnetic field was generated by a portable capacitor bank described in Ref. [6]. Our bank consists of a set of dry-film 800-μF 2-kV capacitors (EPCOS, B25690C2807K003) and a 3-kV dc voltage source (Matsusada Precision, HUNS-3P117). The circuit was designed to generate unipolar field pulses. A small coil with an inner diameter of 10 or 6 mm, made with copper-silver alloy wire (φ1.0 mm, 14 turns × 10 layers) fixed with glass-powder-reinforced epoxy, was connected to the capacitor bank. The 10-mm coil can generate 12 T at room

temperature with 900-V charging and 34 T in liquid nitrogen with 1700-V charging, whereas the 6-mm coil can generate 15 T (800 V) at room temperature and 43 T (1700 V) at 77 K. The maximum field is limited by the Joule heating of the coil. After charging the capacitors, the coil current is generated by activating the thyristor with a trigger pulse. The circuit is shunted by a switch (not shown) after the generation of the field pulse is completed. The repetition rate of field pulses is limited by various factors. For pulses close to the highest charging voltages mentioned above, it is safe to wait for approximately 10 min after each pulse to avoid accumulation of Joule heating. For smaller pulses, we can achieve a repetition rate of around 2–3 min, which is limited by time consumption of other processes such as data saving, capacitor charging, etc.

As shown in Fig. 2, time-series data of the photodetector signal $V_{PD}(t)$, together with the current-sensor output, the magnetic-field pickup coil output, and the voltage for the phase modulation from the function generator (Siglent, SDG2082X), were collected using a 12-bit digital oscilloscope (Rigol, DHO1204 or DHO5108). Data acquisition was triggered by the 5-V pulse that was also used to trigger the output of the pulse-magnet current. Data collected for about 10 ms with a time interval of 1 or 2 ns amounted to approximately 500 MB.

IV. EVALUATION OF MAGNETO-OPTICAL ROTATION ANGLE

For ordinary steady-state MOKE measurements using the loopless Sagnac interferometer, lock-in amplifiers are used to extract the MOKE signal from the photodetector output [10,15,16,18,19]. In contrast, for MOKE measurements under pulsed magnetic fields, it is difficult to use lock-in amplifiers, because their time constants must be much shorter than the pulse width. In fact, we tried using a digital lock-in amplifier (NF Corporation, LI5660). However, it turned out that the time constant had to be as short as 1 μ s to avoid artificial delay of the response, while the noise and higher-harmonic rejections were not sufficient with such a short time constant.

Thus, we decided to use numerical lock-in analysis to obtain the MOKE signal from the time-dependent photodetector voltage signal. In this work, we developed formulas to perform the numerical lock-in analysis with proper phase handling. Analyses based on these equations enable us to determine both the amplitude and sign of the n th harmonic signals in the photodetector voltage. Details of this phase-resolved lock-in analysis are described in Appendixes C1–C3.

From the evaluated in-phase signals of the n th harmonic signal $V_{in,n}$, we obtain the total MO rotation signal θ_{tot} , which consists of the sample Kerr angle θ_K and background signal θ_{bk} originating from Faraday rotation of optical components, using the relation

$$\theta_{tot} = \theta_K + \theta_{bk} \simeq \frac{1}{2} \frac{J_2(2\phi_m)}{J_1(2\phi_m)} \frac{V_{in,1}}{V_{in,2}}, \quad (1)$$

where $J_n(x)$ is the n th order Bessel function of the first kind. For this relation, it is optimal to use the modulation depth ϕ_m of around 0.92 rad, which corresponds to the maximum in $J_1(2\phi_m)$. For different modulation depths ϕ_m , this relation can

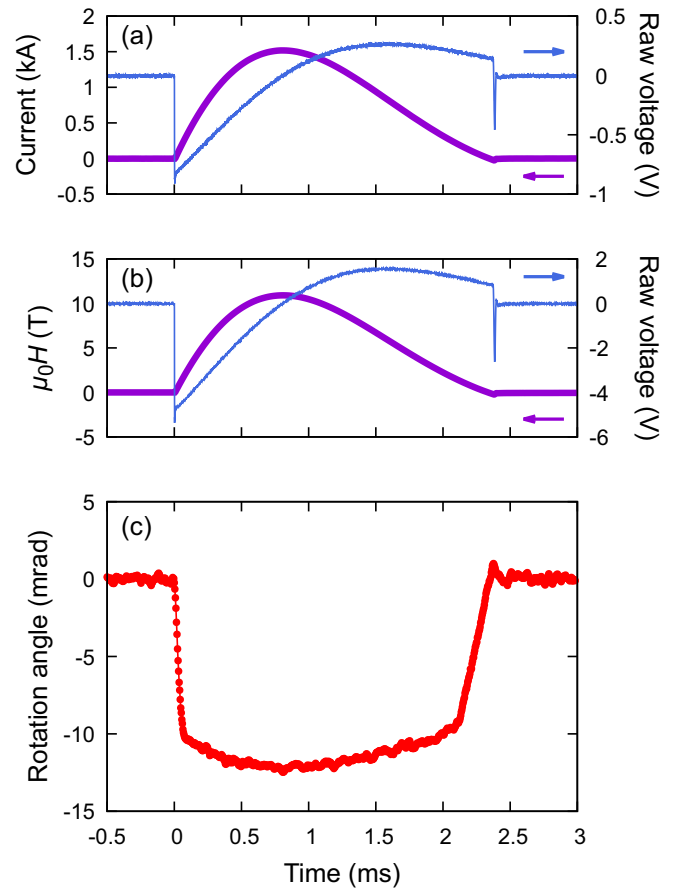


FIG. 3. Typical voltage signals of (a) current and (b) field sensors recorded by the oscilloscope (blue curves; right vertical axes) for the 10-mm bore coil at room temperature. The current and field values evaluated by numerical integration of the sensor voltages are shown with purple curves (left vertical axes) in panels (a) and (b), respectively. (c) Time dependence of the magneto-optical rotation signal measured with Fe evaluated using numerical lock-in technique based on Eq. (3).

be generalized as

$$\theta_{tot} \simeq \frac{1}{2} \frac{J_{m_e}(2\phi_m)}{J_{m_o}(2\phi_m)} \frac{V_{in,m_o}}{V_{in,m_e}}, \quad (2)$$

where m_e and m_o are certain even and odd integers. We can also use another convenient relation

$$\theta_{tot} \simeq \frac{V_{in,3}}{V_{in,2} + V_{in,4}} \sqrt{\frac{15V_{in,2} + 24V_{in,4} + 9V_{in,6}}{80V_{in,4}}}, \quad (3)$$

when we use ϕ_m of around 2.1 rad [i.e., corresponding to the peak in $J_3(2\phi_m)$]. The proof of this relation is given in Appendix C3. This equation is useful because one does not need to know ϕ_m very accurately before each measurement. This advantage would be important in large-scale pulse-field experiments, where machine time is often quite limited.

A concrete example of the data analysis process is described in detail in Appendix C4.

V. RESULTS AT ROOM TEMPERATURE

In Fig. 3, we plot representative time-dependent data obtained with the 10-mm bore coil at room temperature. The current and field sensor voltages are shown by the blue curves in panels (a) and (b). Since these voltages are, respectively, proportional to $-dI/dt$ and $-dH/dt$, we can obtain coil current and magnetic field by numerically integrating the sensor voltages and then multiplying by appropriate geometry factors. The resulting $I(t)$ and $\mu_0 H(t)$ curves are shown with the purple curves, demonstrating successful generation of a field pulse with 2-ms duration. The resolution of the magnetic-field sensor (pickup coil) is 0.002–0.003 T, which is primarily attributed to electrical noise. In addition, the resolution is also limited by the temporal resolution (13 μ s for the data shown in this paper, as discussed in Appendix C). Within 13 μ s, the magnetic field changes by 1% of the pulse height in the beginning of the pulse.

In the 2-ms pulse, we succeeded in obtaining the MO rotation angle θ_{tot} , as shown in Fig. 3(c), by using the numerical lock-in analysis described in Sec. IV. This curve was obtained using pure Fe (Nilaco, 99.99% purity). When the pulse starts at $t = 0$, a steep change in θ_{tot} due to ferromagnetic domain alignment was observed. Subsequently, a small and gradual modulation in θ_{tot} was observed, which is attributed to the Faraday-effect background θ_{bk} , as discussed in the next paragraph. Near the end of the pulse, a large change in θ_{tot} was again observed, due to the randomization of ferromagnetic domains in the sample. Since the coercivity of pure Fe is known to be very small, θ_{tot} almost returns to zero when the field pulse finishes. Notice that the field changing rate is not uniform within a pulse [see, e.g., Fig. 3(b)]. Since the field changes fastest in the beginning of the pulse, the change in θ_{tot} is also fastest there, whereas the changes in H and θ_{tot} as a function of time are slower at the end of the pulse. Because of the fast field change, the number of data points per magnetic field is lowest in the very beginning of the pulse, somewhat lowering the temporal and Kerr-angle resolutions. Nevertheless, this issue in measuring MOKE in low-field regimes can be simply overcome by repeating MOKE measurements with pulses of smaller heights, as demonstrated in Fig. 4.

Figure 4(a) shows the field dependence of θ_{tot} for the ferromagnets Fe and Ni (Nilaco, 99 + % purity), as well as nonmagnetic Au (surface of gold-plated brass), measured with pulses of various heights. For Fe, we polished the surface, whereas Ni and Au were used as bought without additional surface treatment. Notice that the “Fe, 800 V” curve is plotted using exactly the same data as those of Fig. 3, without additional averaging or smoothing processes. For clarity, data for Ni are also shown in Fig. 8 in Appendix B with a finer vertical scale. For Ni and Fe, steep changes in θ_{tot} in the low-field region due to saturation of the ferromagnetic moment were observed. Under high fields above the saturation fields, all curves in Fig. 4(a), including that of nonmagnetic Au, exhibit linear field dependences with similar slopes of around -0.2 mrad/T. Above saturation fields, the field dependence of θ_K of a ferromagnet is expected to be very small [21]. Also, the intrinsic Kerr angle of Au is known to be $+0.0093$ mrad/T [22], which is only 1/20 of the observed slope. We have also found that the magnitude of the slope changes when

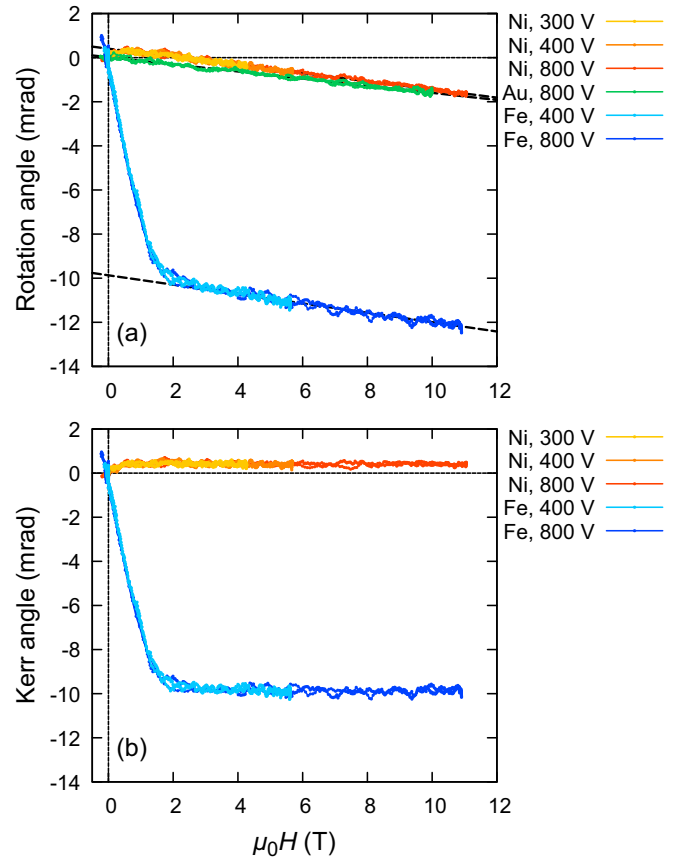


FIG. 4. Results of pulse-field MOKE for various materials at room temperature plotted as a function of magnetic field. The optical power of the light source was 0.8 mW. (a) Total measured MO rotation θ_{tot} containing the Kerr effect from the sample and the background Faraday effects. Here, θ_{tot} was evaluated using Eq. (3). (b) Kerr angle θ_K from the samples obtained by subtracting the field-linear background contribution. For both panels, curves with bluelike colors, orangelike colors, and green represent data for Fe, Ni, and Au, respectively. For Ni and Fe, data obtained with different charging voltages are shown.

we intentionally change the position of the sample fixture inside the coil. Thus, the linear field dependence commonly observed in the total rotation is a background contribution θ_{bk} and is attributable to the Faraday effect from the optical parts (lens, 1/4 wave plate, and fiber). Notice that similar values of the slopes were obtained as long as samples were positioned exactly at the field center, as shown in Fig. 4. This fact indicates the high reproducibility of the setup.

By subtracting θ_{bk} from θ_{tot} , we obtain the Kerr angle shown in Fig. 4(b). For the background subtraction, we assume field-linear background $\theta_{\text{bk}} = \alpha \mu_0 H$ and fit the data under high field with the relation $\theta_{\text{tot}}(H) = \alpha \mu_0 H + \theta_{K\infty}$, where $\theta_{K\infty}$ is the Kerr angle above the saturation field. Subsequently, we subtracted the linear background term from the total rotation $\theta_K(H) = \theta_{\text{tot}}(H) - \alpha \mu_0 H$. From this analysis, $\theta_{K\infty}$ was obtained to be -9.87 ± 0.03 mrad for Fe and $+0.407 \pm 0.01$ mrad for Ni. Overall, the resolution of our experiment is on the order of 0.1 mrad, which is better than previous pulse-field MOKE studies (0.2-mrad resolution in Ref. [8]). The reported values of θ_K at 1550 nm

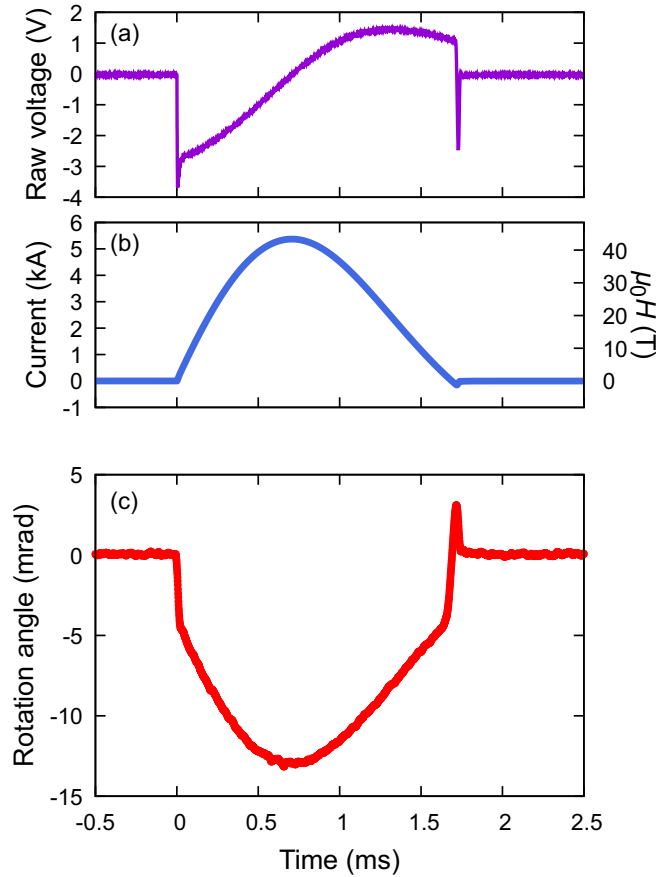


FIG. 5. Time-series data of a pulse-field MOKE experiment over 40 T. The field was generated using the 6-mm coil placed in liquid nitrogen. (a) Voltage signal from the current sensor, recorded by the oscilloscope. (b) Current calculated by numerically integrating the sensor voltage. The corresponding field obtained using a calibrated field-current ratio is shown on the right vertical axis. (c) Time dependence of the magneto-optical rotation signal (including both the sample Kerr rotation and background contribution) measured with Fe_3O_4 using Eq. (1).

wavelength fall within the range $-8.4 \text{ mrad} \leq \theta_K \leq -7.6 \text{ mrad}$ for Fe [23–25] and within the range $+0.31 \text{ mrad} \leq \theta_K \leq +0.46 \text{ mrad}$ for Ni [23,25]. Thus, our observed values of Fe are about 15%–20% larger than the reported values, while that of Ni is in the range of the variation in the reported values.

VI. RESULTS AT LOW TEMPERATURE

In order to demonstrate the applicability of our MOKE technique under more extreme conditions, we performed measurements at 77 K on the (001) surface of a magnetite (Fe_3O_4) single crystal (MTI Corporation). Here, the pulse-field coil was immersed in liquid nitrogen so that we could apply higher current due to lower lead resistance and higher cooling capacity. Note that the sample is also placed directly in liquid nitrogen, whereas the reflection is not significantly affected even in liquid nitrogen owing to the high stability and short focal length of our ferrule-based fixture. The voltage obtained from the current-sensor coil for a 6-mm bore magnet under 1700 V charging is plotted in Fig. 5(a). For the data in Fig. 5,

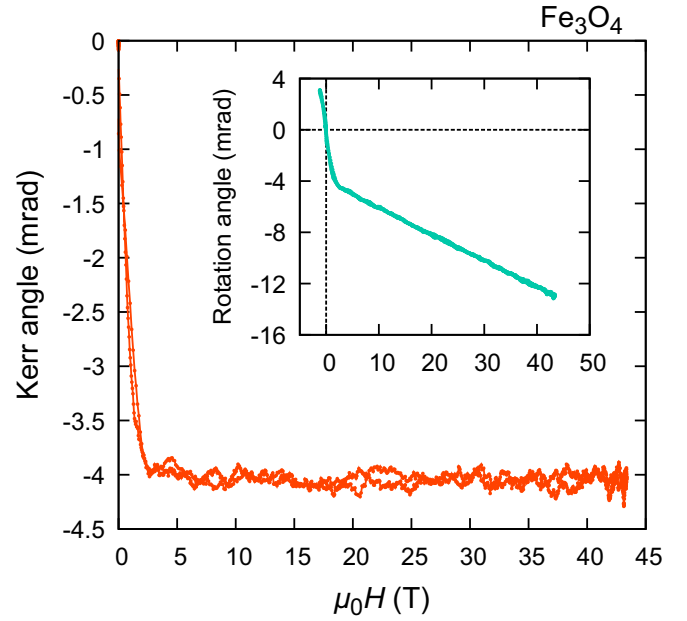


FIG. 6. Demonstration of pulse-field MOKE measurements exceeding 40 T on the (001) surface of magnetite (Fe_3O_4) performed at 77 K. The optical power of the light source was set to approximately 7.5 mW. The total MO rotation θ_{tot} , shown in the inset, is evaluated using Eq. (1). The main panel shows the Kerr angle θ_K of the sample obtained after subtracting the background contribution $\theta_{\text{bk}} = \alpha \mu_0 H$ with a slope $\alpha = -0.205 \text{ mrad/T}$. The saturation value ($-4.048 \pm 0.005 \text{ mrad}$) is in good agreement with a previously reported value [26].

the field value is calculated from current, using a calibration factor between current and field obtained in a prior calibration. As shown in panel (b), the field reached 43 T, which is very close to the maximum dc field achieved at high-field facilities in the world [4]. The time dependence of θ_{tot} plotted in Fig. 5(c) exhibits features similar to the data shown in Fig. 3(c): The steep changes at the beginning and at the end of the pulse correspond to the domain-alignment signal, whereas the gradual changes in the middle of the pulse arise from the background contribution.

In Fig. 6, we show θ_{tot} as a function of magnetic field. As shown in the inset, the raw data exhibit linear field dependence up to 43 T due to the background Faraday contribution with a slope of $\theta_{\text{bk}}/(\mu_0 H) \sim -0.205 \text{ mrad/T}$. The persistence of linear behavior indicates a successful measurement of θ_{tot} up to 43 T. After subtracting θ_{bk} , we obtain θ_K of Fe_3O_4 , as shown in the main panel of Fig. 6. The observed saturation value is $-4.048 \pm 0.005 \text{ mrad}$, which is close to the literature value on the (110) surface (-4.36 mrad at 0.8 eV, room temperature, and 1.76 T) [26], including its sign. The small difference is attributable to anisotropy effects or influence of the charge order (Verwey transition) at around 120 K [27]. The closeness of the experimental and literature values again indicates successful measurements of θ_K . The resolution of this low-temperature measurement was around 0.1 mrad, which equals the resolution achieved in the room-temperature experiment.

To the best of our knowledge, previous reports of pulse-field MOKE are limited to only room-temperature

experiments with maximum fields of up to 11 T [8,9]. Thus, the present study largely exceeds the temperature and field limits of pulse-field MOKE technique, demonstrating the applicability of MOKE in much more extreme conditions than previously recognized.

VII. SUMMARY

In this work, we present a technique for measuring polar MOKE under pulsed magnetic fields. With successful measurements of MOKE above 40 T and down to 77 K with a resolution of 0.1 mrad, we demonstrate that MOKE is indeed a powerful technique to measure magnetic properties under pulsed fields. We also emphasize that our setup is remarkably simple and readily applicable to stronger field apparatus at high-field facilities, including 1000-T destructive magnetic compression magnets [1–3,5]. Moreover, MOKE has a key advantage: it is applicable to small samples such as thin films or nontransparent samples including metallic materials, compared with other magnetic probes such as magnetization measurements or Faraday rotation. Thus, for example, with pulse-field MOKE, we would be able to detect magnetic phase transitions in nontransparent, flakelike samples or thin films, for which bulk magnetization measurements using pickup coils or Faraday rotation measurements are not applicable. Such studies would contribute significantly to the advancement of high-magnetic-field materials science.

In the end, we emphasize that the sample fixture is also applicable to static or zero-field experiments. The fixture enables plug-and-play MOKE experiments free from complicated alignment procedure, resulting in significantly higher experimental throughput. Thus, this paper contributes also to the expansion and acceleration of MOKE studies aimed at investigating nontrivial time-reversal-symmetry breakings in various classes of materials.

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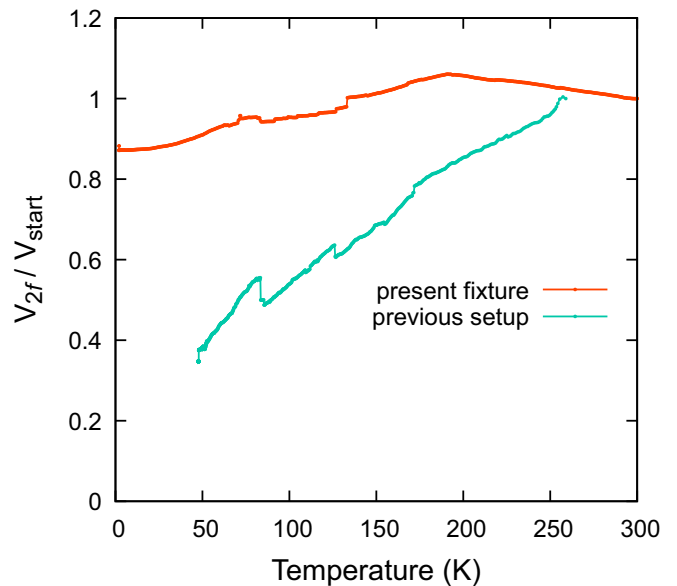


FIG. 7. Example of the change in the reflection during cooling. Here, we plot the amplitude of the second harmonic signal, which is proportional to the power of the reflected light, normalized by its initial value at the start of cooling, V_{start} . These measurements were performed in commercial ^4He cryostats. For the previous setup used in Ref. [17], the reflected power often dropped by several tens of percent upon cooling even if we carefully optimized the reflection at room temperature, as shown by the green curve. In contrast, in the present ferrule-based setup, the decrease of reflection upon cooling is at most around 10% as shown by the orange curve. We also emphasize that this ferrule-based setup eliminates the need for time-consuming optimization before cooling.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

APPENDIX A: TEMPERATURE DEPENDENCE OF THE REFLECTION

Another advantage of the present ferrule-based sample fixture is the stability of the reflection, for example, against mechanical vibration and thermal contraction. Typical temperature dependence of the reflection is presented in Fig. 7. Upon cooling down to 2 K, the intensity varies by only approximately 10% with the present fixture, while the intensity often drops by more than 50% with our previous setup consisting of multiple screws to align the sample [17]. Since the present fixture consists of fewer parts without any moving elements, the fixture is robust against external perturbations.

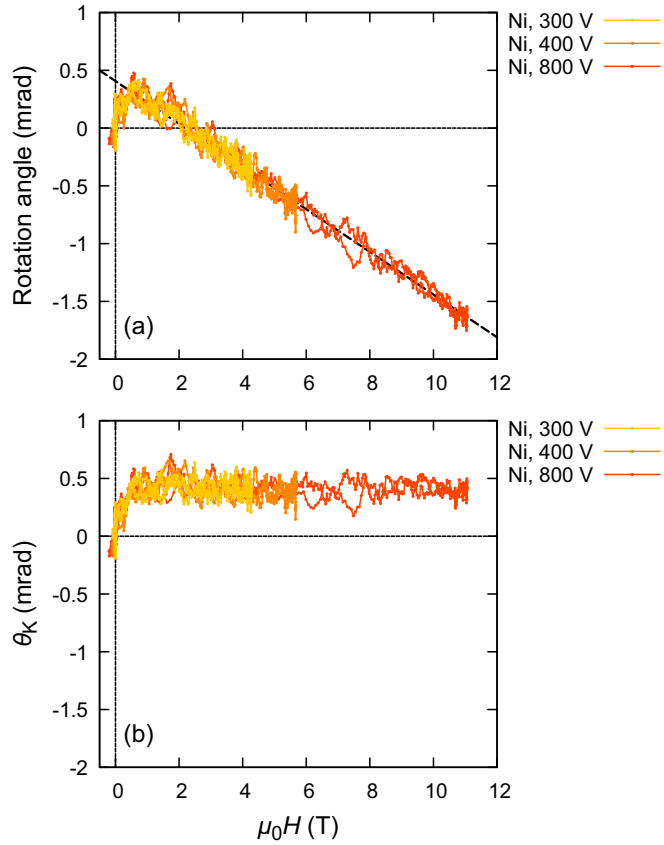


FIG. 8. (a) Raw magneto-optical rotation signal measured with Ni. The sign change around 2 T originates from the cancellation between θ_K and Faraday background contribution. The black dashed line is a fit used to evaluate the background contribution. (b) θ_K obtained after subtracting a linear background. The saturation value is consistent with the previous reports [23,25].

APPENDIX B: DATA FOR NICKEL

We here present graphs showing only results of MOKE measurements on Ni, in order to more clearly display its θ_K , which is smaller than that of Fe. Figure 8(a) shows the field dependence of θ_{tot} measured using pulses of various strengths. The sign change in θ_{tot} observed around 2 T originates from the cancellation between θ_K and θ_{bk} . After subtracting θ_{bk} , a typical ferromagnetic field dependence of θ_K is recovered, as shown in Fig. 8(b).

APPENDIX C: DETAILS OF NUMERICAL LOCK-IN ANALYSIS WITH A PROPER PHASE TREATMENT

1. Numerical lock-in analysis

Here, we begin by discussing numerical lock-in analysis in general. Consider a time-periodic signal of the form

$$V_{\text{sig}}(t) = V_{\text{dc}} + \sum_{i=0} [V_{x,i} \sin(\omega_i t) + V_{y,i} \cos(\omega_i t)].$$

To extract the amplitude and phase of the ω_j component, a reference signal $V_{x,\text{ref}}(t) = V_r \sin(\omega_j t)$ is multiplied by V_{sig} and then is passed to a low-pass filter (LPF) with sufficiently

low cutoff frequency:

$$V_{\text{sig}}(t) \times V_{x,\text{ref}}(t) \propto V_{x,j} \sin^2(\omega_j t) + V_{y,j} \sin(\omega_j t) \cos(\omega_j t) + \text{other oscillatory terms} \xrightarrow{\text{LPF}} \frac{1}{2} V_{x,j}.$$

With this process, we obtain a dc signal proportional to $V_{x,j}$. Similarly, multiplication by $V_{y,\text{ref}}(t) = V_r \cos(\omega_j t)$ and subsequent use of LPF yield a dc component proportional to $V_{y,j}$.

This process is electrically performed in a lock-in amplifier, but it can also be implemented numerically using time-series datasets. One advantage of such a numerical lock-in analysis is that types and parameters of the LPF can be adjusted after taking the data. Moreover, when performing the numerical LPF operation at a certain time t_1 , one can use both the forward and backward data. This substantially reduces artificial response delay. Additionally, we can use many n th harmonic signals to obtain the Kerr angle, possibly enhancing the accuracy of the obtained values [see Eq. (3)].

In this work, we employ simple moving-average LPF with an averaging period of an integer N multiple of the signal period $1/f = 2\pi/\omega$ applied symmetrically to both forward and backward data, i.e.,

$$V_{\text{LPF},x}(t_1) = \frac{f}{2NK} \int_{t_1-N/f}^{t_1+N/f} V_{\text{sig}}(t) \times V_{x,\text{ref}}(t) dt,$$

where K is a scale factor. Compared with other LPFs such as Gaussian filters, this simple moving-average LPF has the advantage of effectively rejecting higher harmonic components [28] even with a relatively short integration time (equivalent to time constant) $\tau \sim N/f$. For the data shown in this paper, we use $N = 20$ for $f = 1.52$ MHz, corresponding to the time constant of $N/f = 13$ μs . This value defines the time resolution of our experiments. This time resolution can be improved by selecting a smaller N at the expense of the signal-to-noise ratio of the MO rotation angle.

2. Phase-resolved analysis of the photodetector signal

In order to evaluate the Kerr angle including its sign, it is necessary to properly account for the phase of the oscillatory signal. For a measurement using the loopless Sagnac interferometer, the photodetector signal $V_{\text{PD}}(t)$ under phase modulation with its depth ϕ_m and angular frequency ω , with consideration of a phase shift due to an arbitrary choice of the origin of the time t_0 , is given by

$$V_{\text{PD}}(t) = V_0 + V_m \cos[2\phi_m \sin(\omega(t - t_0)) + 2\theta_{\text{tot}}],$$

where $\theta_{\text{tot}} = \theta_K + \theta_{\text{bk}}$ is the measured total MO rotation consisting of the sample Kerr angle θ_K and the background contribution θ_{bk} . This expression is rewritten using the Jacobi-Anger expansion:

$$\begin{aligned} V_{\text{PD}}(t) = & V_0 + V_m J_0(2\phi_m) \cos(2\theta_{\text{tot}}) + 2V_m \sum_{n=1}^{\infty} J_{2n}(2\phi_m) \\ & \times \cos(2\theta_{\text{tot}}) \cos[2n\omega(t - t_0)] - 2V_m \sum_{n=0}^{\infty} J_{2n+1} \\ & \times (2\phi_m) \sin(2\theta_{\text{tot}}) \sin[(2n+1)\omega(t - t_0)]. \end{aligned}$$

This can be further rewritten in the form

$$V_{PD}(t) = V_{dc} + \sum_{n=1}^{\infty} [V_{x,n} \sin(n\omega t) + V_{y,n} \cos(n\omega t)],$$

where

$$V_{x,n} = V_n \cos \delta_n, \quad V_{y,n} = V_n \sin \delta_n.$$

Here,

$$V_n = \begin{cases} 2V_m J_n(2\phi_m) \sin(2\theta_{tot}) & (n : \text{odd}), \\ 2V_m J_n(2\phi_m) \cos(2\theta_{tot}) & (n : \text{even}) \end{cases}$$

and

$$\delta_n = \begin{cases} \pi - n\omega t_0 & (n : \text{odd}), \\ \pi/2 - n\omega t_0 & (n : \text{even}) \end{cases}$$

represent the amplitude and phase of the n th harmonic component, respectively, and can be measured using a lock-in amplifiers or by numerical lock-in analysis.

If the phases δ_n are properly evaluated, then

$$\alpha_n = \begin{cases} \delta_n - \pi H[J_n(2\phi_m)] & (n : \text{odd}), \\ \delta_n - \pi H[J_n(2\phi_m)] + \pi/2 & (n : \text{even}) \end{cases} \quad (C1)$$

should be equal to $-n\omega t_0$, and thus α_n should be proportional to n . Here, $H(x)$ is the Heaviside step function [$H(x) = 1$ for $x > 0$ and 0 for $x < 0$], which is used to correct an additional π phase shift that occurs when $J_n(2\phi_m)$ is negative. The relation $\alpha_n = -n\omega t_0$ is used to verify the correctness of the phase evaluation, as well as to obtain ωt_0 from the slope of an α_n - n plot. (For a concrete example, see Fig. 10.) Once ωt_0 is determined, we define the phase-correction angles

$$\tilde{\delta}_n = \begin{cases} \pi H[J_n(2\phi_m)] - n\omega t_0 & (n : \text{odd}), \\ \pi H[J_n(2\phi_m)] - \pi/2 - n\omega t_0 & (n : \text{even}) \end{cases} \quad (C2)$$

in order to obtain the phase corrected in-phase voltage from measured $V_{x,n}$ and $V_{y,n}$ as

$$V_{in,n} = V_{x,n} \cos(\tilde{\delta}_n) + V_{y,n} \sin(\tilde{\delta}_n), \quad (C3)$$

which is further used to determine the Kerr angle in the next subsection. Notice that it is better to use $\tilde{\delta}_n$ determined from multiple harmonic components than to use the measured δ_n , because odd-harmonic signals tend to be very small and thus the measured δ_n for odd n can contain uncertainties.

As a consistency check, it is useful to evaluate the out-of-phase component after phase correction:

$$V_{out,n} = -V_{x,n} \sin(\tilde{\delta}_n) + V_{y,n} \cos(\tilde{\delta}_n). \quad (C4)$$

This quantity should be close to zero if the phase analysis is properly performed.

3. Evaluation of the Kerr angle

In previous MOKE studies using the loopless Sagnac interferometer [10,15,17], θ_{tot} was evaluated from the ratio of the amplitudes of the first and second harmonic signals $|V_1|$ and $|V_2|$ using the relation $|\theta_{tot}| \simeq (J_2(2\phi_m)/2J_1(2\phi_m))(|V_1|/|V_2|)$. The modulation depth ϕ_m should be determined prior to the MOKE measurement, by measuring the dependence of V_2 on the amplitude V_m of the phase modulation signal. Typically, $2\phi_m = 1.84$, which corresponds to the peak of $J_1(2\phi_m)$, is

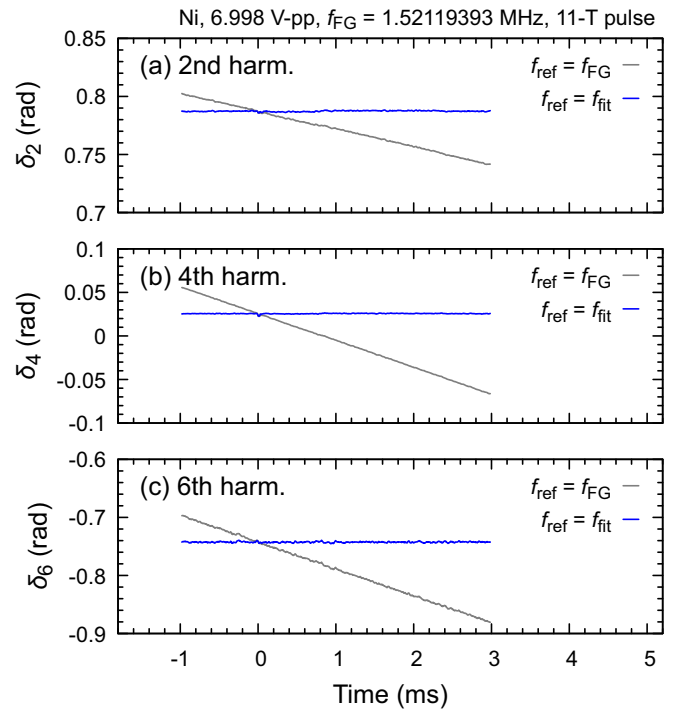


FIG. 9. Comparison of the voltage-signal phase $\delta_n = \arctan(V_{y,n}/V_{x,n})$ for the harmonic components of $n =$ (a) 2, (b) 4, (c) 6 deduced from the numerical lock-in analysis. When using the nominal function-generator output frequency $f_{FG} = 1.521\,193\,93$ MHz as the reference frequency f_{ref} for the numerical lock-in analysis, artificial phase drift occurs due to a small frequency mismatch (gray curves). By instead using the frequency obtained from the fitting of the recorded function-generator signal, $f_{fit} = 1.521\,192\,70$ MHz, the drift is completely suppressed as shown with the blue curves. Note that the difference between f_{FG} and f_{fit} is as small as 1.23 Hz. The absence of the phase drift is crucial for phase-resolved analysis.

used to maximize MOKE sensitivity. This modulation depth yields $J_2(2\phi_m)/J_1(2\phi_m) = 0.543$, and thus we obtain $|\theta_{tot}| \simeq 0.271|V_1/V_2|$.

A very similar relation is also applicable to the phase corrected signals:

$$\theta_{tot} \simeq \frac{1}{2} \frac{J_2(2\phi_m)}{J_1(2\phi_m)} \frac{V_{in,1}}{V_{in,2}}.$$

The advantage of this relation over the previous one is that it allows determination of the sign of the MO rotation angle. We will demonstrate the importance of this capacity in Appendix C 4. For different modulation depths, this relation can be generalized as

$$\theta_{tot} \simeq \frac{1}{2} \frac{J_{m_e}(2\phi_m)}{J_{m_o}(2\phi_m)} \frac{V_{in,m_o}}{V_{in,m_e}},$$

where m_e and m_o are certain even and odd integers, respectively.

We can also use another convenient relation:

$$\theta_{tot} \simeq \frac{V_{in,3}}{V_{in,2} + V_{in,4}} \sqrt{\frac{15V_{in,2} + 24V_{in,4} + 9V_{in,6}}{80V_{in,4}}}.$$

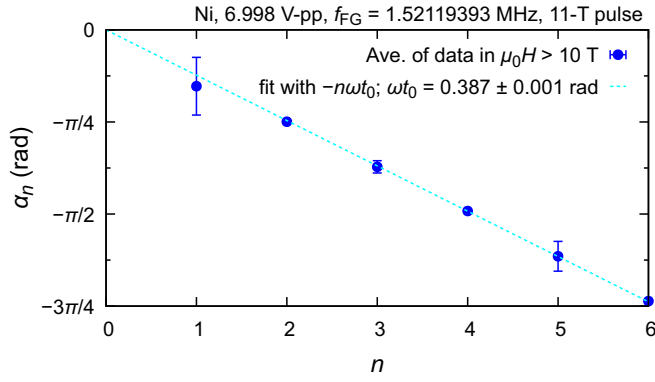


FIG. 10. Phase α_n [defined in Eq. (C1)] of harmonic components of $n = 1$ to 6. Experimental values (blue points) are obtained by averaging phase data above 10 T and the error bars indicate the standard deviation. The data are well fitted by the theoretical relation $\alpha_n = -n\omega t_0$ (light-blue line), yielding $\omega t_0 = 0.387$ rad.

This equation is advantageous because it does not require precise determination of ϕ_m prior to each measurement. This advantage would be valuable in large-scale pulse-field experiments where machine time is often severely limited.

We here provide a derivation of this relation. This is based on the recurrence relation of the Bessel functions:

$$J_{n+1}(x) + J_{n-1}(x) = \frac{2n}{x} J_n(x).$$

This identity leads to various expressions such as $J_2(x) + J_4(x) = 6J_3(x)/x$, $J_3(x) + J_5(x) = 8J_4(x)/x$, and $J_4(x) + J_6(x) = 10J_5(x)/x$. From these, we obtain

$$\frac{J_2(x) + J_4(x)}{6} + \frac{J_4(x) + J_6(x)}{10} = \frac{J_3(x) + J_5(x)}{x} = \frac{8}{x^2} J_4(x).$$

Solving this equation yields

$$x^2 = \frac{240J_4(x)}{5J_2(x) + 8J_4(x) + 3J_6(x)}.$$

This implies that the modulation depth can be obtained by measuring the amplitude of second, fourth, and sixth harmonic components. Furthermore, we find

$$\frac{J_3(x)}{J_2(x) + J_4(x)} = \frac{x}{6} = \sqrt{\frac{20J_4(x)}{15J_2(x) + 24J_4(x) + 9J_6(x)}}.$$

Returning to the relations $V_{in,n} = V_m J_n(2\phi_m) \cos(2\theta_{tot})$ (n : even) and $V_{in,n} = V_m J_n(2\phi_m) \sin(2\theta_{tot})$ (n : odd), the above equation allows us to express the MO rotation angle from second, third, fourth, and sixth harmonic signals by using the relation

$$\begin{aligned} \theta_{tot} &\simeq \frac{V_{in,3}}{V_{in,2} + V_{in,4}} \frac{J_2(2\phi_m) + J_4(2\phi_m)}{2J_3(2\phi_m)} \\ &= \frac{V_{in,3}}{V_{in,2} + V_{in,4}} \sqrt{\frac{15V_{in,2} + 24V_{in,4} + 9V_{in,6}}{80V_{in,4}}}. \end{aligned}$$

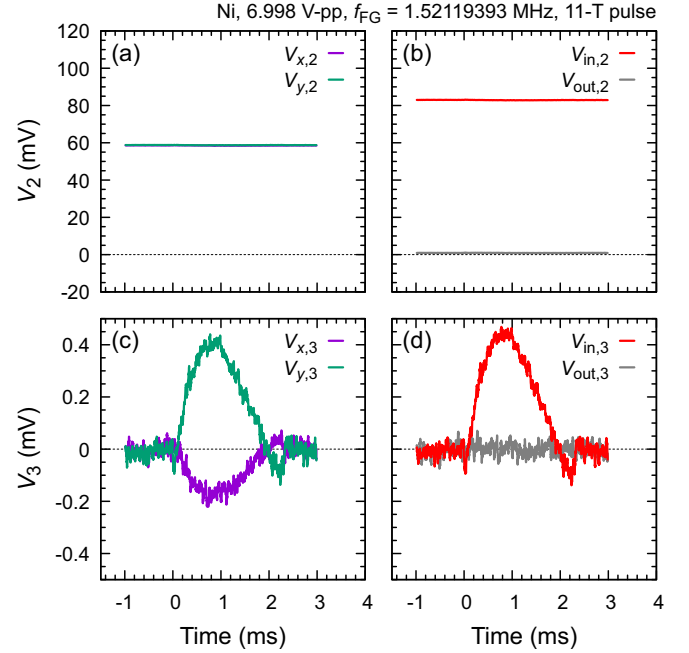


FIG. 11. Comparison of n th harmonic voltages with and without the phase correction. (a) Raw voltages for the second harmonic signal $V_{x,2}$ (purple curve) and $V_{y,2}$ (green curve). (b) In-phase and out-of-phase voltages for the second harmonic signal $V_{in,2}$ (red curve) and $V_{out,2}$ (gray curve) obtained after the phase correction using Eqs. (C3) and (C4) with δ_2 . (c), (d) Similar plots for the third harmonic signal. Notice that V_{out} is close to zero both for the second and third harmonic signals, indicating the validity of the phase-correction analysis. It should also be emphasized that the in-phase signal V_{in} preserves the sign information, in contrast to the absolute value $|V_n|$.

4. Actual process of the analysis

In this subsection, we concretely explain the analysis process by taking the MOKE measurement of Ni foil (Nilaco, 0.05-mm thickness, 99+% purity) under an 11-T pulse field as an example. The frequency of the phase modulation signal output from the function generator (FG) was determined to be $f_{FG} = 1.521\,193\,93$ MHz based on prior measurements of the frequency dependence of V_2 using a lock-in amplifier. The amplitude of the phase modulation was 6.998 V-pp, corresponding to the phase modulation depth of $\phi_m = 2.17$ rad.

First, in order to evaluate the phase of the signal, the accuracy in the frequency of the reference signal is very important. There exists small but noticeable difference in the actual frequency and the nominal frequency f_{FG} . If we use f_{FG} in the numerical lock-in analysis, even a slight frequency mismatch leads to artificial phase drift in the lock-in voltages, as illustrated by the gray curves in Fig. 9. Thus, we recorded the FG signal by the oscilloscope, as shown in Fig. 2, and fitted it with a sinusoidal function to evaluate the frequency f_{fit} with an accuracy better than 0.01 Hz. In the present case, f_{fit} differs from f_{FG} only by 1.23 Hz. Nevertheless, this small correction eliminates the phase drift entirely, as shown by the blue curves in Fig. 9. This absence of the phase drift is crucial for performing the following phase-resolved analysis.

From the phases of the even-order harmonic components, as well as the odd-order components under high magnetic

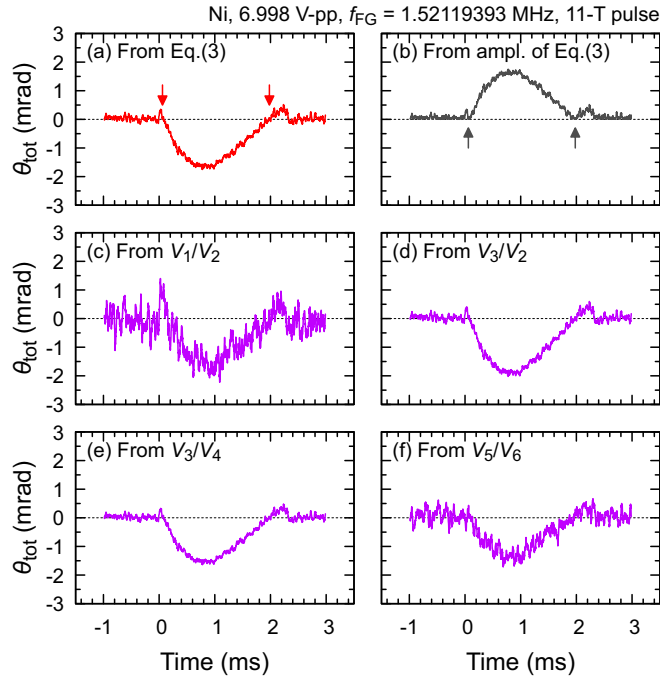


FIG. 12. Comparison of the raw polarization rotation angle θ_{tot} evaluated using various equations. (a) θ_{tot} obtained using Eq. (3). (b) Incorrect evaluation of θ_{tot} using Eq. (3) but based only on the amplitude information. As indicated by the arrows in panels (a) and (b), the sign change occurring due to the competition between sample and background signals observed in panel (a) is lost in panel (b), demonstrating the importance of the phase-resolved analysis. θ_{tot} from ratios (c) V_1/V_2 , (d) V_3/V_2 , (e) V_3/V_4 , and (f) V_5/V_6 . The method using Eq. (3) [panel (a)] is consistent with these conventional evaluations [panels (c)–(f)].

fields, we evaluate α_n [defined in Eq. (C1)], which is plotted as a function of the harmonic number n in Fig. 10. All data points lie on a single straight line, consistent with the theoretical relation $\alpha_n = -n\omega t_0$. From the slope of this plot, we evaluate ωt_0 as 0.387 rad and subsequently calculate δ_n using Eq. (C2).

We finally obtain the phase-corrected in-phase voltage $V_{\text{in},n}$ using Eq. (C3), as exemplified in Fig. 11. Notice that, although $V_{\text{in},n}$ resembles the absolute value $|V_n|$, the former has an important advantage that it preserves the sign information. Therefore, this phase-resolved analysis is essential for evaluating the sign of the MOKE signal. It is also worth mentioning that the out-of-phase component $V_{\text{out},n}$ is close to zero for all n , as predicted by the theory, further supporting the validity of the analysis.

We compare the total MO rotation angle θ_{tot} evaluated using various relations in Fig. 12. The result obtained from Eq. (3) with phase-resolved analysis is shown in Fig. 12(a). For comparison, we plot an incorrect result obtained from the same equation but only from the amplitude information in Fig. 12(b). As indicated by the arrows in panels (a) and (b), the sign changes occurring at 0.06 and 2.00 ms, originating from a cancellation between θ_K and θ_{bk} , are replaced by extrinsic dips in the analysis without the phase consideration. This result highlights the importance of phase-resolved analysis explained in Appendix C 2. In Figs. 12(c) and 12(d), we plot θ_{tot} evaluated by simple ratios between odd and even harmonic in-phase signals [Eq. (2)]. In these evaluations, we use the nominal modulation depth $\phi_m = 2.17$ rad determined from modulation depth sweep measurements. The result obtained by Eq. (3) agrees well with those from the simple evaluations, manifesting the validity of the relation. Since the modulation depth is optimized for the use of V_3 , the noise levels in panels (c) and (f) are relatively high.

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