Micro-MOKE with optical interference in the study of 2D \( \text{Cr}_2\text{Ge}_2\text{Te}_6 \) nanoflake based magnetic heterostructures

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ABSTRACT
Due to its high sensitivity and sub-micrometer spatial resolution, the microscopic magneto-optical Kerr effect (micro-MOKE) technique has been widely utilized in the study of two-dimensional (2D) magnetic materials and heterostructures. Here, by taking 2D \text{Cr}_2\text{Ge}_2\text{Te}_6 nanoflakes on a silicon wafer substrate as a model system, we present the effect of optical multilayer interference induced “optical artifacts” on the effective micro-MOKE measurements in such a system. It is found that not only the magnitude but also the sign of the micro-MOKE signal could be modulated by the “optical artifacts” with the variation of either the \text{Cr}_2\text{Ge}_2\text{Te}_6 nanoflakes’ thickness or probe light wavelength. The detailed analysis based on the multilayer interference model reveals that there are two kinds of MOKE signals, sign reversal and magnitude modulation behavior, and the interference effect on the MOKE response can be easily predicted from unpolarized optical images. Our findings provide instructional principles on the promotion of micro-MOKE characterization as well as the optical studies in van der Waals magnets.

Recently, two-dimensional (2D) van der Waals magnets, for example \text{Cr}_2\text{Ge}_2\text{Te}_6 (CGT) and \text{CrI}_3, are drawing tremendous attention due to their remarkable physical properties as well as potential applications in the minimization and optimization of spintronic devices. With the reduction in their thicknesses down to few or even single atomic layer, the effective probing of the magnetization and its dynamics in those 2D magnets become a big challenge. Several devices, such as the superconducting quantum interference device (SQUID) and magnetic force microscope (MFM), and techniques, such as the anomalous Hall effect (AHE), have been utilized to study the magnetization of 2D magnets. Among them, the microscopic magneto-optical Kerr effect (micro-MOKE), which refers to the polarization plane rotation of a linearly polarized light when reflected by magnetized materials, is widely used as a highly sensitive method with a theoretical probing limit of $10^{-12}$ emu. By focusing the probe light down to the diffraction limit, submicrometer spatial resolution can be achieved. Moreover, micro-MOKE measurements do not require physical contacts between the probe and the sample, which allows for the existence of additional capping layers on top of the probed area. These features make micro-MOKE a suitable tool for the investigation of thin-layer 2D van der Waals magnets as they are generally embedded between substrates and capping layers and are usually small in physical size (micrometer scale) and in absolute values of magnetization.
Comparing to bulk cases, the micro-MOKE measurements for 2D magnetic nanoflakes are different. When the 2D magnetic crystals are exfoliated into nanoflakes and the thickness of such nanoflakes is reduced below the penetration depth of the probe light, not only the nanoflake itself but also the additional layers and interfaces will play non-negligible roles, for example, leading to extrinsic "optical artifacts" by introducing optical interference. Those "optical artifacts" would not only alter the output MOKE signal but also probably influence the understanding of the magnetic properties of these 2D materials. It is therefore important to explore and to estimate the extrinsic contributions to the overall MOKE response of a multilayer structure composed of a 2D magnetic layer and nonmagnetic dielectric layers, especially when strict numerical calculations are difficult to implement for lack of relevant parameters.

In this letter, the "optical artifacts" introduced by multilayer optical interference on the effective micro-MOKE signal was studied in 2D CGT based heterostructures. The CGT thickness ($t_{CGT}$) and probe wavelength ($\lambda$) dependences of the sign and magnitude of the Kerr rotation ($\theta_K$) were found. Within a multilayer interference model, the extrinsic contribution of the optical interference effect on the micro-MOKE response of the multilayer structure is revealed, and a fast and easy approach based on the microscopic image analysis is proposed to predict the interference induced sign changing of the MOKE signal.

Here, the PMMA/CGT/SiO$_2$/Si heterostructure was chosen as a prototype structure which is made by the mechanical exfoliation method based on bulk CGT crystals. The CGT nanoflakes with various thicknesses ($t_{CGT}$) were characterized by an atomic force microscope (AFM). The results can be found in the supplementary material. The micro-MOKE hysteresis loops (obtained in polar geometry at 11 K) of these CGT nanoflakes were measured at three different probe wavelengths (i.e., $\lambda = 532$ nm, 633 nm, and 780 nm) and are shown in Fig. 1. All these loops show distinct ferromagnetic features, and the magnitude and sign of the Kerr rotation angle ($\theta_K$) exhibit significant dependence on $t_{CGT}$ as well as on $\lambda$. For instance, a reverse of the loop was observed at $\lambda = 532$ nm and 780 nm with the increase in $t_{CGT}$ from 22.2 nm to 35.7 nm. Besides, both the sign reversal and magnitude variation of $\theta_K$ took place when $\lambda$ increased from 532 nm to 780 nm for $t_{CGT} = 22.2$ nm and 26.2 nm although the shape of the hysteresis loops remains unchanged for a certain $t_{CGT}$. It is obvious that the measured MOKE signals are modulated by the extrinsic "optical artifacts."

In order to present this further, the $\theta_K$ values at positive saturation magnetic fields are summarized in Fig. 2 where different variation tendencies of $\theta_K$ against $t_{CGT}$ can be found for different $\lambda$. Specifically, when $\lambda = 532$ nm, $\theta_K$ jumps abruptly from $-4$ mrad to $3.3$ mrad within a quite narrow range of $t_{CGT}$ (from 22.2 nm to approximately 25 nm) and then decreases slowly to $3.2$ mrad at $t_{CGT} = 58.3$ nm. A similar tendency can be found in the $\lambda = 633$ nm curve, despite that the sign of $\theta_K$ does not change in the whole range of $t_{CGT}$ and that its magnitude is larger. However, for $\lambda = 780$ nm, $\theta_K$ changes gradually from $-1.1$ mrad to $1.2$ mrad in the 25.5 nm–37.7 nm range of $t_{CGT}$ and keeps nearly constant all along to $t_{CGT} = 58.3$ nm. Therefore, these three curves can be roughly classified into two groups. Both the $\lambda = 532$ nm and $\lambda = 633$ nm curves exhibit an abrupt jump and a following slow declining process with the increase in $t_{CGT}$, while the $\lambda = 780$ nm curve shows a much gentler variation and the smallest magnitude of $\theta_K$.

To better understand the mechanism behind the $t_{CGT}$ and $\lambda$ dependent MOKE response, we modeled our sample with a...
four-layer heterostructure, as shown in Fig. 3(a). The transparent PMMA layer is treated as semi-infinite because the reflectivity at the air-PMMA interface is small and almost independent of $t_{CGT}$ and $\lambda$. When a linearly $p$-polarized beam ($E_{inc,p}$) is normally incident onto the heterostructure, the $p$-polarized reflection ($E_p$) inside the PMMA layer can be expressed as $E_p = E_{p1} + E_{p2}$, where $E_{p1}$ and $E_{p2}$ are interfering $p$ waves reflected by the CGT layer and the SiO$_2$-Si interface, respectively. Similarly, the $s$ component ($E_s$) induced by the magneto-optical effect of the magnetized CGT layer is composed of two interfering $s$ waves ($E_s = E_{s1} + E_{s2}$) that are generated inside the CGT layer. It should be noted that “$p$-” and “$s$-” refer to the initial polarization direction and the polarization of the generated light component due to the magneto-optical effect, respectively.

The Kerr rotation angle $\theta_K$ can be obtained roughly as $-|E_s|/|E_p|$. Its magnitude is closely related to the relative amplitude between $E_s$ and $E_p$, while its sign depends on whether $E_s$ and $E_p$ are in phase or out of phase. Any factors (such as $\lambda$ and $t_{CGT}$) that can modulate the relative amplitude and phase retardation between $E_s$ and $E_p$ are able to affect the magnitude and sign of $\theta_K$. For a certain heterostructure with fixed $t_{CGT}$, its optical property and the interference effect are $\lambda$ dependent. Consequently, the relative amplitude and phase retardation between $E_s$ and $E_p$, and thus the magnitude and sign of $\theta_K$, are also $\lambda$ dependent, which has been proved by our experimental results shown in Figs. 1 and 2.

Similarly, the variation in transmission and reflection properties of the CGT layer with $t_{CGT}$ will also induce significant changes in $\theta_K$ even when $\lambda$ is fixed. In detail, a thin CGT layer with a small $t_{CGT}$ value reflects only a fraction of the incident light and allows the dominant part to transmit through it and get reflected at the SiO$_2$-Si interface ($E_{p1} < E_{p2}$); however, the situation will get reversed ($E_{p1} > E_{p2}$) when $t_{CGT}$ is large enough to block most of the incident light. A similar reversal process will also happen to $E_{s1}$ and $E_{s2}$ when $t_{CGT}$ changes. Such processes will not only modify the amplitudes of $E_p$ and $E_s$ but also change their phase retardation, leading to a magnitude modulation and sign reversal in $\theta_K$. There are two cases where the sign of $\theta_K$ can be altered. In the first case, as shown in Fig. 3(b), when $E_s$ reverses from negative to positive and grows further but $E_p$ does not change much, the Kerr rotation $\theta_K$ will follow the variation of $E_s$ as a result of their proportional relationship. In the second case, if $E_s$ stays almost constant but $E_p$ changes its sign, then $\theta_K$ will jump abruptly across zero and vary inversely in

![Fig. 3](https://example.com/fig3.png) (a) Modeled four-layer structure of the sample and the light path inside. The widths of the arrows are qualitative representation of the corresponding light intensities. Schematic diagrams (b) and (c) represent sign reversal and magnitude modulation processes of $\theta_K$ induced by $E_s$ and $E_p$, respectively.
magnitude with $E_p$ [Fig. 3(c)]. It is easy to find from Fig. 2 that the $t_{CGT}$ dependent $\theta_K$ variation at $\lambda = 780$ nm belongs to the first case, while the variations at 532 nm and 633 nm can be classified into the second one.

To check the wavelength dependent interference further, the phase differences between interfering beams and their cosine values were calculated, as presented in the supplementary material. It is found that for a $\sim$296 nm SiO$_2$ layer, the s waves ($E_{s1}$ and $E_{s2}$) at $\lambda = 532$ nm and 633 nm are in constructive interference, while the p waves ($E_{p1}$ and $E_{p2}$) are in destructive interference; however, the situation is opposite when $\lambda = 780$ nm. As the phase reversal of $E_p$ (or $E_s$) can only happen as a result of the relative amplitude flip between destructively interfering waves $E_{p1}$ and $E_{p2}$ (or $E_{s1}$ and $E_{s2}$), this calculation result is in good accordance with the model analysis and experimental results mentioned above. It confirms the validity of this multilayer interference model and suggests the important role played by optical interference induced “optical artifacts” in the micro-MOKE measurements of such 2D magnetic heterostructures. This kind of “optical artifact” can be adopted to greatly enhance the weak MOKE signal from atomically thin magnets,\textsuperscript{16–18} i.e., by making use of destructive interference to reduce the p-polarized reflection as much as possible; one can improve the proportion of the s-polarized component in the signal beam and consequently enhance the detected $\theta_K$. For example, the reflectivity of a series of CGT nanolakes at $\lambda = 633$ nm is lower than that at $\lambda = 532$ nm, as illustrated in the supplementary material, that explains why the $\lambda = 633$ nm curve always lies above the $\lambda = 532$ nm curve in Fig. 2. This is quite similar in principle to the enhancement of visibility of atomically thin 2D materials deposited on silicon wafers with oxidation layers.\textsuperscript{19,20}

In fact, the interference effect in the MOKE response of magnetic heterostructures has been well studied experimentally and theoretically by many groups.\textsuperscript{17,18,21–23} Now, one can use the optical transfer matrix method to calculate the MOKE signal from a certain layered heterostructure as a function of the probe wavelength and film thicknesses, however, only if the refractive index and thickness of each layer is exactly known.\textsuperscript{24} This is sometimes hard to achieve, especially when new materials are used in the heterostructure. Here, we will present a fast and easy approach based on the microscopic image analysis to predict the interference induced sign changing of the MOKE signal.

The brightness in the microscopic images is proportional to the p-polarized reflection intensity, so it can be used as a measure of $|E_p|^2$. A color image recorded by a digital camera is generally composed of three brightness images corresponding to red (R), green (G), and blue (B) channels, respectively. Among them, the G and R channels correspond roughly to the 532 nm green light and 633 nm red light used in this paper. Therefore, we extracted the G and R channel images from a color image of a typical CGT nanolake and present them in Figs. 4(a) and 4(b) to qualitatively show the G channel brightness (red curve) declines step by step, but the R channel brightness (green curve) decreases first above $t_{CGT} = 23.5$ nm and then increases slightly at $t_{CGT} = 22.2$ nm. It is obvious that the reflected light intensity reaches minimum when $E_p$ is about to reverse its phase because of the resonance cancellation between $E_{p1}$ and $E_{p2}$. The tendencies of image brightness variation vs $t_{CGT}$ indicate that a phase reversal of $E_p$ (thus a sign change in $\theta_K$) took place when $t_{CGT}$ is near 23.5 nm for $\lambda = 532$ nm instead of 633 nm, which agrees with our experimental observations (Fig. 2). This image analysis provides a fast and easy approach to determine the $t_{CGT}$ interval in which $E_p$ (and thus $\theta_K$) is about to reverse its sign. This does not require a magnetized state of the magnetic layer or a full knowledge of the optical parameters of the heterostructure. One can simply use an unpolarized optical image taken at room temperature to do this analysis and will get reliable results since all the contributions (even the effect of the capping layer) are automatically taken into account. Moreover, it is possible to achieve even better results at other wavelengths by using proper monochrome light sources to illuminate the sample when taking images of nanolakes.

In summary, the optical interference induced “optical artifact” effect on the micro-MOKE response has been studied by using a CGT nanolake based 2D magnetic heterostructure as a model system. The sign reversal and magnitude modulation of $\theta_K$ have been observed with variations of either the flake thickness or probe wavelength. By utilizing a multilayer interference model, the “optical
artifact” induced sign reversal and magnitude modulation of θK have successfully been understood. Besides, based on the optical image analysis, we proposed a simple but useful method for predicting the 2D magnetic layer thickness dependence of θK’s sign reversal behavior.

See the supplementary material for the thickness and reflectivity characterization of the CGT nanoflakes and the heterostructure, the micro-MOKE measurements, and the determination of the interference type by phase difference calculation.

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