Development of a tomographic Mueller-matrix scatterometer for nanostructure metrology

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In this paper, we describe the development of a novel instrument, tentatively called tomographic Mueller-matrix scatterometer (TMS), which enables illuminating sequentially a sample by a plane wave with varying illumination directions and recording, for each illumination, the polarized scattered field along various directions of observation in the form of scattering Mueller matrices. The incidence angle is varied from 0° to 65.6° with the rotation of a flat mirror that changes the position of the focal point of a light beam on the back focal plane of a high numerical aperture objective lens. The scattering Mueller matrices are collected over a wide range of scattering angles (0°–67°) and azimuthal angles (0°–360°). The developed instrument was then applied for the measurement of nanostructures in combination with an inverse scattering problem solving technique. The experiment performed on a periodic nanostructure preliminarily demonstrates the performance of TMS as well as its potential in nanostructure metrology. It is expected that the TMS would be a powerful tool for characterizing the polarized scattered-field distributions and measuring nanostructures in nanomanufacturing. Published by AIP Publishing. https://doi.org/10.1063/1.5034440

I. INTRODUCTION

Optical scatterometry, 1–6 also referred to as optical critical dimension metrology, has become one of the most important techniques for measuring the critical dimension (CD) and overlay of nanostructures in semiconductor manufacturing due to its inherent noncontact, nondestructive, time-effective, and relatively inexpensive merits over other metrology techniques, such as scanning electron microscopy (SEM) and atomic force microscopy. It should be noted that the scatterometry described here is different from the traditionally known scatterometry technique. The latter is commonly employed to analyze scattered light from irregular defects and particles aiming to quantify the size and frequency of these random features, while the scatterometry described here is essentially a diffraction-based metrology technique, where the diffracted light from periodic features such as gratings is analyzed to reconstruct structural profiles of these features. Different from the conventional image-based metrology techniques, the measurement in optical scatterometry is not straightforward and typically involves the solution of an inverse problem by fitting the measured data with a multiparameter model that describes the light-nanostructure interaction. Nevertheless, it is not restricted by the well-known Abbe diffraction limit as encountered in image-based techniques and thus plays an important role in addressing devices with sub-wavelength feature sizes in semiconductor industry. At the early stage of optical scatterometry, reflectometry was employed to collect the signature (i.e., reflectance or transmittance) of a sample. 1,2 Since the year of around 2000, spectroscopic ellipsometry (SE) was introduced into optical scatterometry, 3–5 which was traditionally used to characterize thicknesses of thin films and optical constants of both layered and bulk materials according to the change in polarization states of light upon light interaction with a sample. 7 Among the various types of ellipsometers, Mueller matrix ellipsometer (MME), also known as Mueller (matrix) polarimeter, can provide all 16 elements of a 4 × 4 Mueller matrix, which contains all information regarding the polarization properties of a sample. Consequently, MME-based scatterometry, also called Mueller matrix scatterometry, can acquire much more useful information (e.g., anisotropy and depolarization) about the sample than conventional SE and thereby can achieve better measurement sensitivity and accuracy. 8–10

Along with the advantages of optical scatterometry, there are some challenges or limitations to this technique. 11,12 First, the parameter correlation deserves special attention. Large parameter correlation increases the measurement uncertainty and makes the solution of the inverse problem apt to fall into local minima. The parameter correlation issue becomes worse in advanced technology nodes (22 nm and beyond) with the transition from conventional scaling-driven planar devices, e.g., complementary metal-oxide semiconductor (CMOS), to complex three-dimensional (3D) transistor architectures, e.g., fin field-effect transistors (FinFETs), since more parameters must be solved for these 3D features (at least 12 parameters are required for FinFETs, whereas 5 or 6 parameters for planar CMOS). Even worse, as nodes progress and devices shrink, the parameter correlation issue usually compounds with the loss of sensitivity to some parameters. Second, optical scatterometry collects reflected (or transmitted) light from the spot illuminated at a sample surface and delivers it to the detector system. The reconstructed structural profiles are the average results over all features within the illumination spot.

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Any sample structures that are smaller than the spot size will not be accurately discriminated. Thus, optical scatterometry is mostly suitable for measuring periodic dense structures while infeasible for the measurement of isolated or the general non-periodic structures since the light-sample interaction for isolated or non-periodic structures is no longer a simple diffraction problem but a general scattering problem and the collection of scattering information in a specific direction (like the usually adopted specular reflection in optical scatterometry) is inadequate to resolve these features.

In the reported literature, several designs with the philosophy of trying to collect the scattering information as much as possible have been presented, which might be able to address the above challenges or limitations in conventional optical scatterometry. The goniometric optical scatter instrument (GOSI) was developed to achieve in-plane and out-of-plane ellipsometric scattering measurements by combining an ellipsometer system with a goniometer and was applied for the characterization of nanoparticles and rough surfaces. In a single measurement, the GOSI can collect the scattering information at a single scattering direction. To address the scattered-field distribution, GOSI needs to perform measurements at multiple scattering directions in and out of the incidence plane via a rotatable mechanical arm. The goniometric setup was introduced into a deep ultraviolet scatterometer for the measurement of deep sub-wavelength Si (silicon) line structures, which showed sensitivities for nanometer-sized detailed structural parameters such as the corner rounding. Compared with the lensless goniometric setup, a more convenient approach to collect the scattering information is to make a combination with optical microscopy techniques, such as angle-resolved Mueller polarimetry, through-focusing scanning optical microscopy (TSOM), scatterfield microscopy, and tomographic diffractive microscopy (TDM). The angle-resolved Mueller polarimetry was developed by combining MME with a wide-field microscope, which was capable of acquiring the full polarization properties of a sample by imaging the back focal plane (BFP) of a high numerical aperture objective lens (OL). It should be noted that the angle-resolved Mueller polarimetry is also mostly suited to characterize periodic dense structures under a specular reflection configuration. When the sample is an isolated or non-periodic structure, the polarized scattering information is overlapped at the BFP since the sample is illuminated simultaneously under all possible incidence angles in a wide-field microscope. The TSOM provides a strategy for the measurement of isolated or non-periodic structures with a wide-field microscope by analyzing not only the best-focus image but also a set of out-of-focus images of the sample. Another possible strategy is to decouple the overlapped scattering information at the BFP by illuminating the sample under various incidence angles in a sequential manner, as did in scatterfield microscopy and TDM. Compared with the scatterfield microscopy, which only provides the reflectance of a sample, the TDM can record both the amplitude and phase of the field scattered by a sample for different illumination directions. It was demonstrated that a resolution of about a quarter of the illumination wavelength could be achieved with TDM by further making use of the polarization information in the scattered field.

Inspired by the tomographic setup in TDM and considering the rich polarization information contained in a Mueller matrix, we develop a novel instrument in this work, which we tentatively call the tomographic Mueller-matrix scatterometer (TMS). Basically, a TMS experiment consists in illuminating sequentially a sample by a plane wave with varying illumination directions and recording, for each illumination, the polarized scattered field along various directions of observation in the form of scattering Mueller matrices. Specifically, to realize the scanning of the illumination directions, a rotatable flat mirror (FM) is employed in the developed instrument to change the position of the focal point of the light beam on the BFP of a high numerical aperture OL. An epi-illumination setup is designed to collect the scattered-field distribution associated with each illumination by imaging the BFP of the OL. Since each point on the BFP is represented by a Mueller matrix, the full polarization properties of the scattered field are thus achieved. To reconstruct the geometrical profile of the nanostructure under test, an inverse scattering problem is solved by matching the experimental scattering Mueller matrices with the theoretically calculated scattering Mueller matrices.

In the remainder of this paper, we first present the design and operation principle of TMS in Sec. II. Then, we introduce the calibration details for TMS in Sec. III. Since the focus of this paper is on the development of a novel instrument, the TMS, a periodic nanostructure (a Si grating) is chosen to examine the potential of TMS in nanostructure metrology without losing the above focus. Accordingly, the experimental details on the measurement of the investigated sample by TMS, including sample description, data analysis, and experimental results, are introduced in Sec. IV. Finally, we draw some conclusions and show the outlook of the present technique in Sec. V.

II. INSTRUMENTATION

A. Instrument setup

An overview of the TMS is illustrated in Fig. 1. A laser-driven light source (LDLS Eq-99XFC, Energetiq Technology, Inc., USA) connected with a 105 μm diameter optical fiber is employed as the light source. The light beam from the fiber becomes a parallel beam after passing through a collimating lens Lc. The parallel light beam passes successively through a bandpass filter F (FL05632.8-3, Thorlabs, Inc., USA), a flat mirror FM, a polarizer P, the 1st rotating compensator C1, a non-polarizing beam splitter (BS), and a lens L1 and then focuses on the BFP of a high numerical aperture OL (Epiplan-Apochromat 50×/0.95, Carl Zeiss, Inc., Germany). The emerging parallel light beam from the OL illuminates a sample placed at the front focal plane of the OL.

The incidence angle θ is varied with the rotation of a flat mirror by changing the position of the focal point of a light beam on the BFP of the OL, as depicted in the dashed box in Fig. 1. Since the flat mirror is mounted on a high-precision rotating stage (SGSP-60YAW, Sigma Koki, Co.,
Ltd., Japan), the incidence angle can be accurately controlled. This illumination setup for controlling the incidence angle is a well-established method known as the Köhler illumination. The sample is placed on another same rotating stage to change the azimuthal angle $\phi$ of the sample. After passing through BFP which contains the scattered-field distribution of the sample, the image on the BFP is acquired by a sCMOS camera. The focal planes of $L_1$ and $L_3$ are conjugate with the focal planes of $L_2$ and the detecting plane of the camera, respectively. The infinity space between $L_1$ and $L_3$ is able to accommodate necessary optical components. Although the Fourier image that contains the scattered-field distribution can offer a rotation speed of 4400 rpm. The camera is a high-performance of up to 100 frames per second (fps) at full frame.

\section*{B. Instrument principle}

In the TMS, the modulation and demodulation of polarization states are based on a dual rotating-compensator configuration which consists of a fixed polarizer (P), the 1st rotating compensator (C$_{r1}$), a sample (S), the 2nd rotating compensator (C$_{r2}$), and a fixed analyzer (A). The 1st and 2nd compensators are synchronously rotating at $\omega_1 = 5\omega$ and $\omega_2 = 3\omega$, where $\omega$ is the fundamental mechanical frequency. The state of polarization of an input light beam is described by its Stokes vectors $\mathbf{S}_m$. The Stokes vector $\mathbf{S}_{\text{out}}$ exiting from the analyzer is a dot product of the total Mueller matrices of the configuration with the input Stokes vectors $\mathbf{S}_m$. The irradiance signal at each pixel of the camera is proportional to the first element of $\mathbf{S}_{\text{out}}$ and can be expressed as the time dependent waveform due to the continuous rotation of both compensators. The theoretical waveform is given by

\begin{equation}
I(t) = I_0 m_{11} \left\{ a_0 + \sum_{n=1}^{16} \left[ a_{2n} \cos(2n\omega t - \varphi_{2n}) + b_{2n} \sin(2n\omega t - \varphi_{2n}) \right] \right\} = I_0 \left\{ 1 + \sum_{n=1}^{16} \left[ a_{2n} \cos(2n\omega t - \varphi_{2n}) + b_{2n} \sin(2n\omega t - \varphi_{2n}) \right] \right\},
\end{equation}

where $I_0$ is the spectral response function, $m_{11}$ is the $(1, 1)$ element of the Mueller matrix of the sample, $\varphi_{2n}$ is the angular phase shift, and $\{I_0 = I_0 m_{11} a_0, (a_{2n} = a_{2n}/a_0, b_{2n} = b_{2n}/a_0)\}$ are the d.c. and dc-normalized a.c. Fourier coefficients, respectively. The sample Mueller matrix elements $m_{ij}$ $(i, j = 1, 2, 3, 4)$ are linear combinations of $a_{2n}$ and $b_{2n}$.

For an experimental dual rotating-compensator system, the irradiance measured at every pixel of the camera is expressed as

\begin{equation}
I(t) = I'_0 \left[ 1 + \sum_{n=1}^{16} \left( a'_{2n} \cos 2n\omega t + b'_{2n} \sin 2n\omega t \right) \right],
\end{equation}

where $\{I'_0, (a'_{2n}, b'_{2n})\}$ are the experimental d.c. and dc-normalized a.c. Fourier coefficients. The relationship between
\{I'_0, (\alpha'_n, \beta'_n)\} and \{I_0, (\alpha_{2n}, \beta_{2n})\} obeys the following expression:

\[
I'_0 = I_0, \quad (3a)
\]

\[(\alpha_{2n}, \beta_{2n})]^T = \Re(\varphi_{2n})[\alpha'_n, \beta'_n]^T, \quad (3b)
\]

where the superscript "T" represents the matrix transpose and \(\Re(\varphi_{2n})\) is a \(2 \times 2\) rotation transformation matrix. To detect the waveform given in Eq. (2), one can perform \(K\) times the number of integrals of the irradiance over the fundamental optical period of \(\pi/\omega\), which leads to raw flux data \(\{g_k, k = 1, 2, \ldots, K\}\) of the form

\[
g_k = \int_{(k-1)\pi/K\omega}^{k\pi/K\omega} I'_0 \left[ 1 + \sum_{n=1}^{16} (\alpha_{2n}' \cos 2n\omega t + \beta_{2n}' \sin 2n\omega t) \right] dt
\]

\[
= \frac{\pi I'_0}{K\omega} + \sum_{n=1}^{16} \frac{I'_0}{n\omega} \frac{\sin n\pi K}{K} \left[ \alpha_{2n}' \cos \left( \frac{2k-1)n\pi}{K} \right) + \beta_{2n}' \sin \left( \frac{2k-1)n\pi}{K} \right) \right]. \quad (4)
\]

Since the Fourier coefficients \(\{(\alpha'_n, \beta'_n), n = 9, 12, 14, 15\}\) all vanish, there are only 25 unknowns in Eq. (4), including 24 nonzero Fourier coefficients along with \(I'_0\). Since the highest-order nonzero Fourier coefficient is at 32 rad/s, at least 33 integrations are required over the fundamental optical period to obtain all 25 nonzero Fourier coefficients \(\{I'_0, \alpha'_n, \beta'_n\}, n = 1, 2, \ldots, 8, 10, 11, 13, 16\) by solving Eq. (4). According to Eq. (3), the Fourier coefficients \(\{I_0, \alpha_{2n}, \beta_{2n}\}\) can be further obtained, from which we can finally obtain the sample Mueller matrix elements \(m_{ij} (i, j = 1, 2, 3, 4)\) associated with each pixel of the camera. The Mueller matrices associated with all pixels of the camera compose the scattering Mueller matrices of the sample.

In the common setup of the TMS, the integration time is set as 20 ms and the number of integrals of the irradiance \(K\) is equal to 50 in the fundamental optical period \((\pi/\omega)\). The 1st and 2nd compensators rotate synchronously at \(\omega_1 = 5\pi\) rad/s (2.5 Hz) and \(\omega_2 = 3\pi\) rad/s (1.5 Hz), respectively. To improve the signal-to-noise ratio, the measured flux data \(g_k\) are averaged over 10 fundamental optical periods. Thus, the Mueller matrix measurement at a single incidence angle will take \(\sim 10\) s.

**III. CALIBRATION**

**A. Calibration of incidence angle**

Polarimetric data analysis requires a precise knowledge of the incidence angles during sample measurements. In the developed instrument, the incidence angle is varied with the rotation of a flat mirror by changing the position of the focal point of a light beam on the BFP of the OL. When the sample is illuminated at a normal incidence, the corresponding angle of the flat mirror is set as the original position. Assume that the spherical aberration of \(L_1\) and OL has been well corrected, and according to Abbe’s sine condition, the relationship between the incidence angle \(\theta\) and the rotation angle \(\alpha\) of the flat mirror satisfies

\[
\frac{\sin(\theta)}{\sin(2\alpha)} = \frac{f_{L_1}}{f_{OL}}, \quad (5)
\]

where \(f_{L_1}\) and \(f_{OL}\) are the focal lengths of \(L_1\) and OL, respectively. In practical experiments, the rotation angle \(\alpha\) is adjusted by a precision motor stage with a uniform increment. The incidence angle \(\theta\) was calibrated by using a one-dimensional grating with a period of 1.530 \(\mu m\), and the calibration was performed at the wavelength of 632.8 nm. Figure 2(a) shows the Fourier image of the grating obtained by imaging the BFP of the OL at a normal incidence angle. As can be observed, five bright regions lie in a straight line with an equally spaced distance \(d\) on the BFP, which represent the five \((\pm 2\text{nd}, \pm 1\text{st}, 0\text{th})\) propagating diffraction orders of the reference grating. The 0th order is at the center of the BFP due to the normal-incidence configuration. The distance \(d\) has a linear relationship with respect to \(\sin(\theta)\), with \(\theta\) being the diffraction angle estimated by the well-known grating equation. The angular distance corresponding to a single pixel is thus given by

\[
\Delta \sin(\theta) = \frac{\sin(\theta)}{d}, \quad (6)
\]

where the value of \(d\) is represented by the number of pixels on the camera. On the BFP, when the incidence angle varies, these diffraction orders keep the same space distance with their positions translated together. We chose the 0th order and obtained the shifts of its position at multiple incidence angles. As shown in Fig. 2(b), when there is a small rotation angle \(\Delta \alpha\), the position of the 0th order will move several pixels away from the center of the BFP. Let us denote the moving distance as \(\Delta d\).

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**FIG. 2.** Schematic for calibrating the incidence angle with a known reference grating. (a) The Fourier image of the grating measured by TMS at a normal incidence angle; (b) the position of the 0th diffraction order shifts with the rotation of the flat mirror; (c) \(\sin(2\alpha)\) is plotted against \(\sin(\theta)\) by a linear fit, which leads to a coefficient of determination of \(R^2 = 0.9992\).
Thanks to the specular reflection of the 0th order, the variation of the sine of the incidence angle \( \theta \) will be

\[
\Delta \sin(\theta) = \Delta \sin(\theta) \times \Delta d. \tag{7}
\]

A plot of \( \sin(\theta) \) as a function of the sine of double rotation angle \( \sin(2\alpha) \) of the flat mirror is presented in Fig. 2(e). A linear polynomial fitting was performed, which yielded the slope, intercept, and coefficient of determination \( (R^2) \) of 34.63, −0.0055, and 0.9992, respectively. A good agreement with the linear relation given by Eq. (5) can be easily observed from Fig. 2(c). Several data points slightly deviated from the linear fit might be induced in the determination of the normal incidence angle and the rotating errors of the motor stage. In addition, due to the limited numerical aperture of the employed OL, the maximum value of \( \sin(\theta) \) is about 0.9105, which results in a maximum achievable incidence angle of 65.6° for the TMS.

**B. Calibration of polarization components**

1. **Without the objective lens**

The TMS is based on a dual rotating-compensator configuration, whose calibration parameters include the determination of the actual transmission-axis orientations \( P \) and \( A \) of the polarizer and analyzer, the initial fast-axis orientations \( C_{S1} \) and \( C_{S2} \) of the two compensators as well as their wavelength-dependent phase retardances \( \delta_1 \) and \( \delta_2 \). The calibration of TMS was performed with a standard reference sample (130 nm SiO\textsubscript{2}/Si thin film) by removing the BS, the OL, and other lenses \( L_1-L_3 \). In order to reduce the nonuniform effect of light intensity, the camera was used as a single point detector by averaging the intensities obtained by all the pixels during the calibration. We used a nonlinear regression method to obtain the calibration values of \( P, A, C_{S1}, C_{S2}, \delta_1, \) and \( \delta_2 \). To implement the regression calibration, the sample Mueller matrices were calculated according to Eqs. (2)–(4) and Fresnel’s equations \(^2\) as functions of the calibration parameters and denoted as \( M_{ij}^{\text{calc}}(P, A, C_{S1}, C_{S2}, \delta_1, \delta_2) \). The calculated Mueller matrices were then fitted to the measured Mueller matrices \( M_{ij}^{\text{meas}} \) by adjusting the calibration parameters \( P, A, C_{S1}, C_{S2}, \delta_1, \) and \( \delta_2 \) to minimize a \( \chi^2 \) error function defined as

\[
\chi^2 = \sum_{i,j=1}^{4} \left\{ \frac{m_{ij}^{\text{meas}} - m_{ij}^{\text{calc}}(P, A, C_{S1}, C_{S2}, \delta_1, \delta_2)}{\sigma(m_{ij})} \right\}^2, \tag{8}
\]

where \( \sigma(m_{ij}) \) is the standard deviation of the experimental Mueller matrix element. The minimization of \( \chi^2 \) can be done by an iterative nonlinear regression analysis, such as the Levenberg-Marquardt (LM) algorithm.\(^{27}\)

Although the employed BS was declared to be nonpolarizing, it was found that the s- and p-polarizations after the BS still had a minor shift with each other in both transmission and reflection setups. The residual polarization effect of the BS will be incorporated into the measured sample Mueller matrices and affect the measurement accuracy. To further refine the calibration, we characterized polarization properties of the BS before inserting it into the TMS. The polarization properties of the BS in transmission and reflection setups were represented as the beam splitter reflection matrix \( M_{bs}^{t} \) and the beam splitter reflection matrix \( M_{bs}^{r} \), respectively. The \( M_{bs}^{t} \) and \( M_{bs}^{r} \) were measured by using the well-calibrated dual rotating-compensator system, which were then directly used to correct the residual polarization effect of the employed BS in actual experiments.

2. **With the objective lens**

The polarization effect of the high numerical aperture OL also provides a challenge to the measurement accuracy and shall be corrected prior to the experiment. Generally, the Mueller matrix \( M_{ob} \) measured by the TMS consists of five parts, which can be written as

\[
M_{ob} = M_{bs}^{t} \cdot M_{ob}^{id} \cdot M_{id} \cdot M_{id}^{*} \cdot M_{bs}^{r}, \tag{9}
\]

where \( M_{ob}^{id} \) and \( M_{id}^{*} \) are the Mueller matrices of the OL in an illumination direction and a collection direction, respectively, and \( M_{id} \) is the Mueller matrix of a sample. As shown in Eq. (9), to obtain the sample Mueller matrix \( M_{id} \), it is required to determine the Mueller matrices \( M_{bs}^{t} \) and \( M_{bs}^{r} \) of the BS as well as \( M_{ob}^{id} \) and \( M_{ob}^{cd} \) of the OL. The calibration of \( M_{bs}^{t} \) and \( M_{bs}^{r} \) has been described in Sec. III B 1. To calibrate \( M_{ob}^{id} \) and \( M_{ob}^{cd} \) of the OL, a spherical mirror was employed as the reference sample.\(^{28}\) In the calibration, we assumed that the constituent materials of the OL were reciprocal and isotropic. The calibration of \( M_{ob}^{id} \) and \( M_{ob}^{cd} \) was performed in two different setups.

Figure 3(a) illustrates the setup for the calibration of \( M_{ob}^{id} \) in the illumination direction. As shown in this figure, a parallel light beam focuses on the BFP of the OL after passing through \( L_1 \) and then parallelly illuminates on the spherical mirror at a certain incidence angle. Due to an extremely small illumination spot size (~100 µm), the reflection on the surface of the spherical mirror can be approximated to be a plane reflection. By overlapping the front focal plane of the OL with the diametral plane of the spherical mirror, the incidence light returns back along the same path and focuses on the BFP. The focal point on the BFP is then imaged on the camera by other lenses. Since the focal point on the camera consists of several pixels, the intensities associated with the adjacent pixels are averaged to estimate the Mueller matrix. The measured Mueller matrix in this case can be represented by

\[
M_{ob} = M_{bs}^{t} \cdot M_{ob}^{id} \cdot M_{id} \cdot M_{id}^{*} \cdot M_{bs}^{r}, \tag{10}
\]

![FIG. 3. Schematic for calibrating the polarization effect of the employed objective lens. (a) Setup for the illumination direction. (b) Setup for the collection direction. Other optical components that are not presented in this figure are assumed to be well aligned to image the BFP on the camera.](image-url)
FIG. 4. The calibrated Mueller matrices (normalized to $m_{11}$, which is not shown) of the employed objective lens in the illumination direction at different incidence angles and the wavelength of 632.8 nm. The incidence angles are calibrated using the method in Sec. III A with a maximum of 65.6°.

where the sample Mueller matrix $M_s = \text{diag}(1, 1, -1, -1)$, which corresponds to the Mueller matrix for reflection at normal incidence.\cite{29} According to Eq. (10), we can obtain $M_{ob}^{id}$ of the OL by

$$M_{ob}^{id} = \left[ M_s^{-1} \cdot (M_{bs}^r)^{-1} \cdot M_m \cdot (M_{bs}^t)^{-1} \right]^{1/2}.\quad (11)$$

Figure 4 presents the calibrated Mueller matrices of the OL in the illumination direction at different incidence angles and the wavelength of 632.8 nm. We can observe from Fig. 4 that $M_{ob}^{id}$ is close to an identity matrix at small incidence angles, implying that the polarization effect of the OL is ignorable. With the increment of incidence angles, an obvious deviation from the ideal identity matrix can be observed from this figure, which suggests that the OL will introduce significant polarization effect in measurements. In practical experiments, the calibrated $M_{ob}^{id}$ can be directly substituted into Eq. (9) to correct the polarization effect of the OL at the corresponding incidence angle.

Figure 3(b) illustrates the setup for the calibration of $M_{ob}^{cd}$ in the collection direction. Since the scattered light emits in a cone and each light ray has an independent illumination direction, we removed $L_1$ to generate a focused light beam with an emitting cone to simulate the same scattering process in actual measurements. As shown in Fig. 3(b), the same spherical mirror is used to determine $M_{ob}^{cd}$ with its center superimposing with the front focal point of the OL. In this case, each illuminating light ray normally emits on the spherical mirror and then reflects back along the same path. By imaging the BFP of the OL, each pixel of the camera with a polar coordinate of $(\theta_s, \phi_s)$ corresponds to a Mueller matrix that can be represented by

$$M_m = M^t_r \cdot M_{ob}^{cd} \cdot M^r_s \cdot M_{ob}^{cd} \cdot M^t_s,$$\quad (12)

where $\theta_s$ and $\phi_s$ are the scattering angle and azimuthal angle of the scattered light, respectively, and the sample Mueller matrix $M_s = \text{diag}(1, 1, -1, -1)$. According to Eq. (12), the $M_{ob}^{cd}$ of the OL can be obtained by

$$M_{ob}^{cd} = \left[ M_s^{-1} \cdot (M^t_r)^{-1} \cdot M_m \cdot (M^t_s)^{-1} \right]^{1/2}.\quad (13)$$

Figure 5(a) presents the calibrated Mueller matrices of the OL in the collection direction. The maximum scattering angle $\theta_s$ is about 67° due to the limitation of the imaging components in TMS, and the azimuthal angle $\phi_s$ of the scattered light is varied from 0° to 360°. As can be observed from this figure, the calibrated Mueller matrices are dependent on azimuthal angles since they are calculated based on the global x-y coordinate system. A rotation matrix must be applied to each pixel of the measured Mueller matrices to rotate them from the x-y coordinate system to the local s-p coordinate system that is commonly adopted to describe polarization by

$$M(s,p) = R(\varphi) \cdot M(x,y) \cdot R(\varphi),$$\quad (14)

where $R(\varphi)$ is the Mueller rotation transformation matrix for rotation by an angle $\varphi$ defined in Fig. 5(a). Figure 5(b) shows the $M_{ob}^{cd}$ of the OL in the s-p coordinate system. A noticeable deviation from the ideal unit matrix can be observed from this

FIG. 5. The calibrated Mueller matrices of the employed objective lens in the collection direction. (a) and (b) represent the Mueller matrices of the objective lens in the x-y and s-p coordinate systems, respectively. The wavelength and maximum scattering angle in the calibration are 632.8 nm and 67°, respectively.
figure at large scattering angles, which may induce significant measurement errors in experiments. In addition, the two $2 \times 2$ diagonal blocks of the Mueller matrices exhibit a rotational symmetry, which is in accordance with the expectation of a radially dependent birefringence and dichroism of the OL. In practical experiments, the calibrated $M^{cal}_{ij}$ is substituted into Eq. (9) to correct the polarization effects of different pixels on the BFP of the OL.

IV. EXPERIMENT

A. Sample description

A Si grating is investigated to examine the potential of the developed TMS. The Si grating was fabricated by e-beam lithography followed by dry etching and was used as a template in nanoimprint lithography. Figure 6 shows the cross-sectional SEM micrograph of the grating structure as well as the adopted geometrical model to characterize its profile. As shown in Fig. 6, the geometrical profile of the Si grating is characterized by top critical dimension $x_1$, grating height $x_2$, and sidewall angle $x_3$. The period of the Si grating is 800 nm. The nominal dimensions of other structural parameters of the Si grating are $x_1 = 350$ nm, $x_2 = 470$ nm, and $x_3 = 88^\circ$, respectively. In the solution of the inversion problem, we fixed the grating pitch at 800 nm and just let the parameters $x_1 - x_3$ vary. The optical constants of the Si substrate were fixed at values taken from the literature. The Si grating was chosen for the study mainly based on the following considerations. First, the Si grating had been fully investigated in the previous literature using conventional Mueller matrix scatterometry, which can thereby provide a more objective estimation to the measurement precision and accuracy of the developed instrument in nanostructure metrology. Second, the scattering problem for a periodic nanostructure degrades to a relatively simple diffraction problem, which is beneficial for us to examine the capability of TMS in the collection of the scattered-field distributions.

B. Data analysis

Scattered-field distributions of a sample are measured with the TMS at multiple incidence angles. For a periodic nanostructure, the scattered field is simply several diffraction orders, and theoretical Mueller matrices of each order can be calculated by rigorous coupled-wave analysis (RCWA). In RCWA, both the permittivity function and electromagnetic fields are expanded into Fourier series. Afterwards, the tangential filed components are matched at boundaries between different layers, and therefore, the boundary-value problem is reduced to an algebraic eigenvalue problem. The overall reflection coefficients can be calculated by solving this eigenvalue problem. According to the reflection coefficients, the $2 \times 2$ Jones matrix $J$ associated with the $n$th order diffracted light of the sample, which connects the incoming Jones vector with the diffracted one, can be formulated by

$$
\begin{bmatrix}
E_{ip} \\
E_{as}
\end{bmatrix} = J
\begin{bmatrix}
E_{ip} \\
E_{is}
\end{bmatrix} = \begin{bmatrix} r_{pp} & r_{ps} \\ r_{sp} & r_{ss} \end{bmatrix}
\begin{bmatrix}
E_{ip} \\
E_{is}
\end{bmatrix}.
$$

where $E_{as}$ refers to the electric field component perpendicular and parallel to the plane of incidence, respectively. In the absence of depolarization, the $4 \times 4$ Mueller matrix $M$ can be calculated from the Jones matrix $J$ by

$$
M = A(J \otimes J^*)A^{-1},
$$

where the symbol $\otimes$ denotes the Kronecker product, $J^*$ is the complex conjugate of $J$, and the matrix $A$ is given by

$$
A = \begin{bmatrix}
1 & 0 & 0 & 1 \\
1 & 0 & 0 & -1 \\
0 & 1 & 1 & 0 \\
0 & 1 & -1 & 0
\end{bmatrix}.
$$

The reconstruction of the nanostructure profile from the TMS-measured scattering Mueller matrices is a typical inverse diffraction problem with the objective of finding a profile whose theoretical scattering Mueller matrices can best match the measured scattering Mueller matrices. A weighted least-squares regression analysis (the LM algorithm) is performed by adjusting the structural parameters under measurement until the calculated and measured data match as close as possible. The solution is obtained by minimizing a weighted mean square error function $\chi^2$ defined by

$$
\chi^2 = \frac{1}{15N - P}\sum_{k=1}^{N}\sum_{ij=1}^{4}\left[\frac{m_{ij}^{meas} - m_{ij}^{calc}(x)}{\sigma(m_{ij})}\right]^2,
$$

where $k$ indicates the $k$th incidence or azimuthal angle from the total number of $N$, indices $i$ and $j$ mean all the Mueller matrix elements except $m_{11}$ (normalized to $m_{11}$), $x$ is a $P$-dimensional vector consisting of the structural parameters under measurement, $m_{ij}^{meas}$ indicates the Mueller matrix elements measured by any pixel of the camera, $m_{ij}^{calc}(x)$ denotes the calculated Mueller matrix elements associated with the vector $x$, and $\sigma(m_{ij})$ is the estimated standard deviation associated with $m_{ij}$. The fitting procedure delivers 95% confidence limits of $1.96 \times \chi^2 \times \sqrt{C_{ii}}$ for the structural parameters, where $C_{ii}$ is the $i$th diagonal element of the structural parameter covariance.

C. Results and discussion

The Si grating was measured by the developed TMS at multiple incidence angles. The measurement wavelength was
632.8 nm with a 3 nm bandwidth, and the illumination spot was about 100 μm. In the experiment, we fixed the azimuthal angle of illuminating light to the Si grating at φ = 0° and varied the incidence angle from 0° to 65.6°. According to the grating equation, we know that the investigated Si grating only has three propagating diffraction orders at the wavelength of 632.8 nm, namely, the 0th and ±1st diffraction orders. However, due to the limitation of the numerical aperture of the employed OL, we could not simultaneously collect all the three diffraction orders on the BFP over the whole range of incidence angles. Thus, only two diffraction orders were used in the following reconstruction of the grating profile. As an example, Fig. 7 shows the scattering Mueller matrices of the Si grating by imaging the BFP of the OL at an incidence angle of 32.3°. As can be observed, there are only two diffraction orders collected by the TMS since the scattering angle of another diffraction order is beyond the maximum collecting angle of the employed OL. The left and right points represent the 0th and ±1st diffraction orders with the corresponding scattering angles of 32.3° and 15.1°, respectively. At the azimuthal angle of φ = 0°, i.e., with the incidence plane perpendicular to grating lines, the two 2 × 2 off-diagonal blocks of the Mueller matrices vanish, as can be observed from Fig. 7, while other elements can be expressed in terms of conventional ellipsometric angles Ψ and Δ, i.e., $m_{12} = m_{21} = -\cos 2\Psi$, $m_{34} = -m_{43} = \sin 2\Psi \sin \Delta$, and $m_{33} = m_{44} = \sin 2\Psi \cos \Delta$ ($m_{11} = m_{22} = 1$). In practice, each diffraction order focused on the camera consists of several pixels, for example, the 0th order consists of 28 pixels. The intensities of the adjacent pixels of the same diffraction order are almost the same. Thus, the Mueller matrix of each diffraction order was obtained by averaging intensities of these adjacent pixels, which was then used to reconstruct the geometrical profile of the Si grating structure.

![FIG. 7. The scattering Mueller matrices of the Si grating on the BFP measured at the wavelength of 632.8 nm. The left point represents the 0th diffraction order at the scattering angle of 32.3°. The right point represents the +1st diffraction order at the scattering angle of 15.1°. The incidence and azimuthal angles of illumination are θ = 32.3° and φ = 0°, respectively.](image)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>SEM</th>
<th>TMS a</th>
</tr>
</thead>
<tbody>
<tr>
<td>x₁ (nm)</td>
<td>350</td>
<td>348.8 ± 0.83</td>
</tr>
<tr>
<td>x₂ (nm)</td>
<td>472</td>
<td>473.4 ± 0.94</td>
</tr>
<tr>
<td>x₃ (deg)</td>
<td>88</td>
<td>87.2 ± 0.14</td>
</tr>
</tbody>
</table>

Table I presents the comparison of fitting parameters obtained from the TMS and SEM measurements, of which the TMS-measured results were extracted from the two collected diffraction orders. According to Table I, we can observe that the TMS-measured results exhibit good agreement with those measured by SEM. A comparison with the previous literature 31,32 also reveals the good agreement between the TMS-measured results and those measured by conventional Mueller matrix scatterometry. Figure 8 shows the fitting results of the measured and calculated best-fit scattering Mueller matrices at multiple incidence angles. Good agreement can be observed from this figure, which yields a fitting error of $\chi^2 = 14.76$. We further calculated the depolarization index distribution associated with the TMS-measured data according to $DI = \sqrt{\text{Tr}((\text{MM})^2) - m_{11}^2}/3m_{11}^2$, $0 \leq DI \leq 1$, 36 where Tr(·) represents the matrix trace. The $DI = 0$ and $DI = 1$ correspond to a totally depolarizing and a totally non-depolarizing Mueller matrix, respectively. The calculated depolarization indices indicated that $|DI - 1| < 0.041$ in the whole range of incidence angles 0°–65.6°. Thus, the depolarization effect could be ignored in the data analysis. We also let the azimuthal angle φ of illumination vary to examine its influence on the final fitting result. The achieved azimuthal angle and the fitting error were $\phi = -0.01° \pm 0.018°$ and $\chi^2 = 14.34$, respectively. It suggested that the increase in fitting parameters did not lead to

![FIG. 8. Fitting result of the measured scattering Mueller matrices and the calculated best-fit scattering Mueller matrices. The incidence angle θ is varied from 0° to 65.6°. The wavelength and the azimuthal angle are fixed at 632.8 nm and φ = 0°, respectively.](image)
a noticeable improvement in the final fitting result. Therefore, the azimuthal angle was fixed at $\phi = 0^\circ$ in the reconstruction. In conclusion, the results presented in Table I and Fig. 8 preliminarily reveal the potential of the developed instrument for accurate nanostructure metrology.

V. SUMMARY AND OUTLOOK

This work presents the development of a TMS for nanostructure metrology. By combining a Mueller matrix ellipsometer with tomographic techniques, the developed instrument enables us to collect the scattering Mueller matrices of a nanostructure in multiple incidence angles. In the calibration process, a regression method has been proposed to calibrate the system parameters. A spherical mirror and a reference grating are used to calibrate the polarization effect of the employed high numerical aperture OL and the incidence angle of the developed TMS, respectively. Some typical specifications of the instrument are summarized as follows. (1) The range of incidence angles is $0^\circ$–$65.6^\circ$; (2) the range of scattering angles is $0^\circ$–$67^\circ$; (3) the range of azimuthal angles of both illuminating and scattered light is $0^\circ$–$360^\circ$; (4) the single measurement time is $\sim 10$ s.

It should be noted that this paper is a report of the first step of the research where the proposal and preliminary verification of the proposed TMS in realizing nanostructure metrology were primarily focused. To provide a more objective estimation to the measurement precision and accuracy of the developed instrument in nanostructure metrology, a periodic nanostructure, namely, a Si grating sample, was chosen for the study since this sample had been fully investigated in our previous work using a conventional Mueller matrix scatterometer.31,32 The comparison between the TMS-measured results and those measured by SEM and the conventional Mueller matrix scatterometer has demonstrated the potential of the developed instrument in nanostructure metrology. Future work will be carried out to investigate the potential of the developed TMS in the measurement of isolated and the general non-periodic nanostructures as well as its detection limit of feature size.

ACKNOWLEDGMENTS

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