

Convenient self-calibrating full-Stokes imaging polarimeter and its application to particle synthesis in reactive plasmas

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Imaging polarimetry is becoming an increasingly significant tool, from remote sensing to material characterization. In reactive plasmas, polarimetry is a well-established *in situ* diagnostic for the size and the complex refractive index of synthesized particles. We present the StokesCam, a self-calibrating, full-Stokes imaging polarimeter based on two micropolarizer cameras. It employs an adapted eigenvalue calibration to address the problem of spatial inhomogeneities and artifacts in polarization optics. A comprehensive uncertainty analysis is provided, using a bootstrapping and Monte Carlo technique to quantify the systematic and statistical errors. The application of the StokesCam to a particle growth process in a reactive plasma is demonstrated as an example of demanding measurement conditions. The device provides high-quality data for scattering modeling and enables not only the spatial monitoring of particle growth but also allows for future studies of the system's asymmetric multiscattering. This work also serves as a practical guide for readers interested in constructing and deploying their own full-Stokes imaging polarimeter.

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I. INTRODUCTION

Polarimetry, hence measuring the wave oscillation properties of light scattered or emitted by a target, is an ever-growing field across multiple disciplines. Emerging from single-beam ellipsometry for the characterization of metals and films in the 19th century [1], the modern mathematical foundation for polarization was built by Stokes [2] and Mueller [3]. Today, the different forms of polarimetry have become standard techniques. They range from simple ellipsometry to complete Mueller polarimeters that can determine the full optical transfer matrices of a sample.

The comparatively modern approach of imaging polarimetry, where the spatial distribution of the polarization signal is measured, has gained increased interest with the availability of high-quality optics and sensor devices.

In life and medical science, imaging polarimetry is increasingly used for structure identification and diagnostics [4–7] to distinguish healthy and cancerous tissue, e.g., of the lung [8] or colon [9]. For that, the birefringence and depolarization properties are useful indicators [10]. They also contain potential information on anomalous glucose concentration, mechanical anisotropy, or density information of the affected tissue [11]. Environmental sciences apply imaging polarimetry for observation through scattering media such as fog or turbid water. The dehazing algorithms can separate the desired background image from the scattering signal based on its polarization [12]. Various polarization 3D reconstruction methods utilize the polarization of light reflected by an object to calculate the 3D position and orientation of the reflective surfaces, resulting in a reconstruction of the shape [13,14]. Single-shot imaging devices are also emerging in ellipsometry [15–17]. Compared with conventional scanning methods, they offer a larger measurement area and an instantaneous surface mapping. This is especially attractive for *in situ* measurements. Another advantage is that imaging devices with appropriate lens systems can be employed at large distances from the target, creating an overlap with the field of remote sensing under ambient light conditions.

Another promising field is the optical *in situ* characterization of particles by polarization-sensitive light-scattering

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techniques. Atmospheric remote sensing is, for example, increasingly relying on polarization information for the analysis and modeling of various aerosols [18], which can otherwise not be readily accessed. When the physical radius a of the scattering aerosols lies in the same order of magnitude as the wavelength λ of the light, the scattering signal becomes very complex to compute due to resonances. For homogeneous, spherical particles, the Mie theory exists as an analytical solution to the scattering problem. It was independently found by Mie [19], Lorenz [20], and Debye [21]. Polarimetry of spherical particles is referred to as Mie-polarimetry [22].

All those applications are reliant on the accurate and complete polarization information, which contains the linear and circular polarization parts as well as any unpolarized fraction. Though the spatial measurement of that is not without challenges, as it cannot be measured at once. Instead, at least four different measurements are necessary [23], each with a different optical setup. For imaging polarimetry, several approaches with varied compromises exist that employ an image sensor [24]. A rather recent development is the reliant micro-polarizer cameras (MPC). These cameras feature arrays of wire-grid polarizers, etched directly onto the imaging sensor [25]. This *division of focal plane* (DoF) approach allows direct measurement of the different linearly polarized fractions of light without further optical equipment, at the cost of spatial resolution. The use of metalenses [26] also falls in that category. Another popular solution is variable components. That can be a rotating wave plate, as used in a rotating-compensator-polarimeter [27,28], or a variable phase retarder [29]. Since several measurements are taken in series and combined into one output image, this is often referred to as *division of time* (DoT). The practical time resolution of those devices is limited, as any interframe motion creates polarization artifacts of some magnitude. Mechanical vibrations of a motor and the temperature dependency of liquid crystal variable retarders can also pose problems in certain environments. A more straightforward approach is *division of amplitude* (DoA), where the necessary measurements are split into independent paths with beam splitters. Although rigid and fast, it results in a decrease in intensity at each detector by a factor of 2.

Another often overlooked obstacle for imaging polarimetry lies in the required calibration. While this is often less of an issue for linearly polarized light, the circular component requires retardation optics, which are wavelength-dependent and exhibit manufacturing-related inaccuracies, limiting the achievable accuracy (see Sec. III A 2). A characterization of all used optical components must also be done in 2D to compensate for any spatial inhomogeneities and even stains on the optics. To further complicate the process, any calibration reference often also exhibits such artifacts. Commercially available calibration targets are also best conditioned for the linear polarization but lack

accuracy for the circular part, introducing systematic errors when used. The sophisticated eigenvalue calibration method (ECM) proposed by Compain *et al.* [30] can mostly overcome this problem, as it does not rely on the precise *a priori* assumptions about the reference components.

Nanodusty plasmas are a complex laboratory system with similar diagnostic requirements to remote sensing. A low-pressure plasma with an admixed reactive gas component, like acetylene or silane, can be used to synthesize nanoparticles. The particles (also called dust) grow from molecular precursors to sizes of several hundred nanometers via a multistage growth process [31]. Polarimetry of scattered light is a well-established diagnostic for such nanoparticle systems [22,31,32]. It not only allows for qualitative *in situ* monitoring of the accretion growth process but also models the particle's time-dependent radius a and their complex refractive index (CRI). Both parameters are crucial for other diagnostics, such as microwave-cavity resonance [33] to determine the dust charge and laser extinction for the dust density [34]. Moreover, precise polarimetry can give valuable insight into the presence of multiscattering or particle size distributions [35]. It, therewith, attends to the stated demand of information-dense *in situ* diagnostics [36].

The modeling challenge lies in the underdetermination of the problem, as the two independent parameters of a polarization ellipse are used to retrieve three values (a and the CRI). One way to resolve the disambiguity is spectroscopic polarimetry, where the polarization signal is measured over a range of wavelengths [37] for each particle size. The approach of kinetic polarimetry, on the other hand, works without spectroscopic equipment. It assumes a sufficiently constant CRI over time and relates the successive measurements of different particle size [38].

Some experimental implementations of polarimetry on nanodusty plasmas exclusively consider the temporal evolution of the linear polarization when accessing the particle radius [39]. This approach requires some strict as well as precise assumptions about the particle's optical properties and an idealized scattering process and geometry. Given the complexity of the scattering problem, this approach is, from our experience, often unsuitable and even prone to creating data artifacts.

A Stokes-complete polarimeter, however, is capable of also measuring the circularly polarized and the unpolarized components. With the additional parameter, the CRI of the particles can be determined using the kinetic Mie approach [38], allowing for a more robust evaluation of the time-dependent particle radius.

Up to the current date, Stokes-complete Mie-polarimetry has almost exclusively been performed as point measurements, although the observed nanoparticle clouds are known to exhibit spatially diverse behavior (e.g., void formation and layered growth patterns). The few exceptions of spatially resolved polarimetry [27,31,40] on

nanodusty plasmas often suffer from low resolution or low light sensitivity.

In this work we bring together the ECM and a modern polarimeter design to present the StokesCam: a full-Stokes imaging polarimeter offering 5MP resolution with a hybrid DoA and DoF setup. Such an approach was first suggested by Shibata *et al.* [41], though, without an appropriate lens system, calibration procedure, or uncertainty discussion. Our device ensures a constant observation angle. In contrast to other popular MPC-based polarimeter designs [42], the proposed setup does not rely on moving or variable components like rotating or liquid crystal wave plates. In total, this makes the StokesCam particularly useful for environments without temperature control or where low vibrations are desired and for precise polarimetric applications in general.

This article also serves as a guide for readers on how to build and deploy their own imaging polarimeter. The target audience is not only researchers in the field of nanodusty plasma, since the presented device and calibration procedure can be widely applied.

The Sec. II offers a brief introduction to the mathematical representation and theory of polarization and its measurement. Sec. III presents the detailed setup of the StokesCam and discusses the necessity and execution of the ECM. Section IV concludes with the application of the StokesCam to a nanodusty plasma environment. The particle size over time is presented as well as the 2-D resolved maps of the polarization signal and indications of multiscattering in the dust cloud.

II. MEASURING POLARIZATION

Before presenting the mathematical foundation for light polarization, a brief overview of the terminology is necessary, considering the various notations used across the multidisciplinary community.

For linear polarization, we define positive rotation angles on a x - y plane as counterclockwise with respect to the x axis, where the z axis coincides with the direction of light propagation. Considering circular polarization, there exist two different reference systems [43] and sign conventions [23], as the derivation from the Maxwell field equations yields an arbitrary definition of the phase difference. We adopt the classical notation that reflects the helicity of the electromagnetic wave. With that, a right-handed circular polarization (*short*: right circular polarization, RCP) means a clockwise rotation of the electric field vector on the plane described above [see also Fig. 1]. In this case, the phase difference Δ of both vector components is defined as positive. Left(-handed) circular polarization (LCP) is given by the counterclockwise rotation.

A. Stokes formalism

Polarization of light is most commonly described by two closely related mathematical formalisms. The Jones

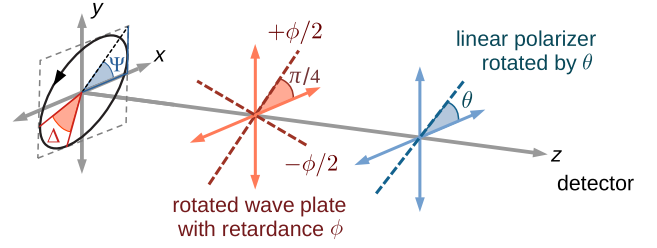


FIG. 1. Minimal polarization measuring setup, consisting of a wave plate with its fast axis oriented at 45° and a linear polarizer (LP) rotated by θ . The polarization ellipse of the incident light is defined by the oscillating electric field vector on a fixed x - y plane. The ellipse can be parametrized by the two ellipsometric angles, where $\tan\Psi$ is the amplitude ratio of the x and y components and Δ their phase difference. The depicted ellipse is defined as left-handed elliptically polarized with respect to the direction of propagation z with $\Delta > 0$.

calculus, based on the complex electric field vector and the more comprehensive Stokes-Mueller formalism. The latter is intensity-based and can account for partial polarization, making it more suitable for real-world applications, where an unpolarized light fraction might be small but inevitable. An in-depth treatise on this matter with the full derivation from the Maxwell equations can be found in Goldstein [23].

In the Stokes-Mueller formalism, the state of a light beam is given by the Stokes vector

$$\mathbf{S} = \begin{pmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{pmatrix} = \begin{pmatrix} I_0 + I_{90} \\ I_0 - I_{90} \\ I_{45} - I_{135} \\ I_{\text{RCP}} - I_{\text{LCP}} \end{pmatrix}. \quad (1)$$

The first element S_0 is the total intensity, whereas the other elements are each given by the predominance between the intensities of two orthogonal polarization states: S_1 and S_2 are the differences of the linear 0° and 90° and, respectively, 45° and 135° polarization directions. S_3 is the difference of the right-polarized to left-polarized intensity.

Each Stokes vector can be separated into an unpolarized S_u and a polarized S_p fraction:

$$\mathbf{S} = \mathbf{S}_u + \mathbf{S}_p = \begin{pmatrix} I_u \\ 0 \\ 0 \\ 0 \end{pmatrix} + \begin{pmatrix} I_p \\ S_1 \\ S_2 \\ S_3 \end{pmatrix}, \quad (2)$$

with I_u being the total unpolarized intensity and I_p the total polarized intensity. The latter is given by the relation

$$I_p^2 = S_1^2 + S_2^2 + S_3^2 \leq S_0^2, \quad (3)$$

bearing a coincident likeness to the physical constraints of a *four-vector* in Minkowski spacetime [44]. The degree of polarization (DoP) can then be defined as

$$\text{DoP} = \frac{\sqrt{S_1^2 + S_2^2 + S_3^2}}{S_0} \leq 1. \quad (4)$$

In combination, one can write the separated Stokes vector as

$$\mathbf{S} = (1 - \text{DoP}) \begin{pmatrix} S_0 \\ 0 \\ 0 \\ 0 \end{pmatrix} + \begin{pmatrix} \text{DoP} \cdot S_0 \\ S_1 \\ S_2 \\ S_3 \end{pmatrix}. \quad (5)$$

When considering polarimetry or ellipsometry, it is useful to rewrite S_p as normalized to the total polarized intensity I_p , because it allows the application of the ellipsometric relations:

$$s_p = \frac{S_p}{\sqrt{S_1^2 + S_2^2 + S_3^2}} = \begin{pmatrix} 1 \\ s_1 \\ s_2 \\ s_3 \end{pmatrix} = \begin{pmatrix} 1 \\ -\cos 2\Psi \\ \sin 2\Psi \cos \Delta \\ -\sin 2\Psi \sin \Delta \end{pmatrix}, \quad (6)$$

where small letters denote the normalized quantities. The angles Ψ and Δ are a parametrization of the polarization ellipse [see Fig. 1] commonly used in the field of ellipsometry. With the Stokes formalism, they can be related to simple intensity-based measurements. Employing the arctan2 function to ensure sign continuity in Δ , the equation becomes

$$\begin{aligned} \Psi &= \arccos(s_1)/2, \\ \Delta &= -\arctan2(s_3, s_2). \end{aligned} \quad (7)$$

Another significant strength of the Stokes-Mueller formalism is its ability to describe any change to the light beam by means of a 4×4 transformation matrix. This Mueller matrix $\mathbf{M}$ represents a physical process or optical element. A setup of successive components is then calculated via left-side matrix multiplications of the corresponding matrices. For example, light with Stokes vector $\mathbf{S}$ that passes through a wave plate and a subsequent linear horizontal polarizer is given by

$$\mathbf{S}' = \mathbf{M}_{\text{LHP}} \cdot \mathbf{M}_{\text{WP}} \cdot \mathbf{S}. \quad (8)$$

Written out as matrices with the respective transmission coefficients $p_{x/y}, r_{x/y} \in [0, 1]$ of both elements and phase retardation ϕ of the wave plate:

$$\mathbf{S}' = \frac{1}{2} \begin{pmatrix} p_x^2 + p_y^2 & p_x^2 - p_y^2 & 0 & 0 \\ p_x^2 - p_y^2 & p_x^2 + p_y^2 & 0 & 0 \\ 0 & 0 & 2p_x p_y & 0 \\ 0 & 0 & 0 & 2p_x p_y \end{pmatrix} \cdot \frac{1}{2} \begin{pmatrix} r_x^2 + r_y^2 & r_x^2 - r_y^2 & 0 & 0 \\ r_x^2 - r_y^2 & r_x^2 + r_y^2 & 0 & 0 \\ 0 & 0 & 2\cos(\phi)r_x r_y & -2\sin(\phi)r_x r_y \\ 0 & 0 & 2\sin(\phi)r_x r_y & 2\cos(\phi)r_x r_y \end{pmatrix} \cdot \mathbf{S} \quad (9)$$

A rotated component is represented via reference-frame rotation by the corresponding angle θ as

$$\mathbf{M}_r = \mathbf{R}(-\theta) \cdot \mathbf{M} \cdot \mathbf{R}(\theta), \quad (10)$$

$$\text{with } \mathbf{R}(\theta) = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos(2\theta) & \sin(2\theta) & 0 \\ 0 & -\sin(2\theta) & \cos(2\theta) & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}. \quad (11)$$

This formalism allows the calculation of arbitrary optical setups if all corresponding matrices are known. A wide range of Mueller matrices for different optical components can be found in standard textbooks, e.g., Goldstein [23].

B. Measuring principle

In principle, the definition in Eq. (1) provides a convenient measurement instruction for the Stokes vector. The components are accessible via an ideal LP (analyzer) and two circular analyzers. These can be realized using a quarter-wave plate (QWP) and a successive LP followed by the transmission axis oriented at $\pm 45^\circ$ relative to the fast axis of the retarder.

In reality, quarter-wave plates are manufactured to work at a specific wavelength. Outside this narrow region, the wavelength-dependent phase shift rapidly differs from the targeted $\lambda/4$. Even for achromatic retarders, the deviation can be several degrees.

This more general case is depicted in Fig. 1: Let the fast axis of an arbitrary retarder be rotated 45° relative to the x - y plane. When polarized light passes the wave plate, it experiences a phase shift ϕ between the orthogonal components. The successive polarizer has a transmission axis that is rotated θ with respect to the reference plane. Executing the Mueller calculus, the intensity I at the detector can then be calculated as

$$\begin{aligned} I_{\theta, \phi} &= \frac{1}{2} [S_0 + S_1 \cos \phi \cos 2\theta + S_2 \sin 2\theta \\ &\quad + S_3 \sin \phi \cos 2\theta]. \end{aligned} \quad (12)$$

This equation has been derived by Stokes [2] in a similar form to originally define the four Stokes parameters in

Eq. (1). Given a suitable set of parameters $\{\phi, \theta\}$, the full polarization information can be gained from a minimum of four measurements. Again, considering the convenient choice of $\theta = 0^\circ, 45^\circ, 90^\circ, 135^\circ$ and under the condition of $\phi \neq n \cdot \pi, n \in \mathbb{N}_{>0}$, the trigonometric terms, they reduce to

$$\begin{aligned} S_0 &= I_{0,0} + I_{90,0} = I_{0,\phi} + I_{90,\phi} = \dots, \\ S_1 &= I_{0,0} - I_{90,0}, \\ S_2 &= I_{45,0} - I_{135,0} = I_{45,\phi} - I_{135,\phi}, \\ S_3 &= \frac{(I_{0,\phi} - I_{90,\phi})}{\sin \phi} - \frac{(I_{0,0} - I_{90,0})}{\tan \phi}, \end{aligned} \quad (13)$$

where S_0 can be gained from any two orthogonal measurements. This conventional method is widely used in polarimetric devices.

Note that accurate results require the precise knowledge of the orientation and especially the phase shift. The inverse tangent in S_3 can grow to a significant contribution when the retardance deviates from $\lambda/4$. While the zero-intensity method employing two crossed polarizers can provide good accuracy for the angular alignment, for the retardance one typically has to rely on manufacturer information (see Sec. III A 2).

As already seen in Eq. (12), the full polarization information cannot be measured with one static setup but instead requires some compromise [24].

III. THE STOKESCAM

The recent advances in the fabrication of polarization-sensitive sensors now offer affordable MPCs [25,45]. These cameras have wire-grid polarizers etched directly onto the imaging chip [Fig. 2(b)]. Each of four adjacent pixels in a 2×2 pattern exhibits a different polarizer direction of $0^\circ, 90^\circ, 45^\circ$, or 135° . Together, they form one superpixel.

Given an observed feature is large compared with the individual pixel size, each superpixel provides the complete linear polarization information. This DoF, however, comes at the expense of spatial resolution.

There exist some prototypes of MPCs with elliptical polarizers using liquid crystal polymer microretarders to capture the full Stokes information [46,47]. Though these devices are rather difficult to manufacture with comparable quality and not as available as linear MPCs.

The following section introduces the StokesCam, a full-Stokes imaging polarimeter. It overcomes the problem of synchronous measurements by employing a split-beam setup first proposed by Shibata *et al.* [41] with two linear MPCs, resulting in a combination of DoF and DoA.

A. Setup

The detailed setup of the StokesCam can be seen in Fig. 2(a). The centerpieces are two *iDS* MPCs of the type *U3-3080 CP-P-GL Rev.2.2*, running a Sony *IMX250MZRC* CMOS sensor [25]. They have 1224×1024 superpixels at a 12-bit monochrome resolution. The $2/3''$ sensor format results in a pixel dimension of $3.45 \mu\text{m}$. Each camera can be triggered externally to ensure frame synchronization. Since a single camera only measures the linear polarization, a nonpolarizing beamsplitter (*Thorlabs BS016*) is used to create two optical paths (DoA). The reflected light is observed by camera B, directly measuring the spatial linear polarization. Because of the reflection, the 45° and 135° components are interchanged, and the image is mirrored. The transmitted beam passes through an achromatic (QWP, *Thorlabs AQWP10M-580*) with the fast axis rotated to 45° . Using the zero-intensity method with an MPC as reference, an alignment error of $\mathcal{O}(0.1^\circ)$ is achievable. The subsequent camera A observes the linear components 45° and 135° that are parallel to the retarder's principal axes. The 0° and 90° pixels contain the information of right and left circular polarization, according to Eq. (12).

As the imaging system, a custom-built bi-telecentric lens is used. It consists of two lenses whose focal planes coincide at an aperture stop, which ideally restricts both entering and exiting rays to those that are parallel to the main axis. This results in a constant observation angle over the image, which is essential when measuring angle-dependent light scattering. Depending on the application, the lens system could also be replaced by a long-distance microscopic lens or a telephoto lens. The telecentricity on the sensor-side ensures that the rays enter the pixel corresponding to the traversed filter. A too-inclined angle of incidence can lead to misregistration [see Fig. 2(b)]. A smaller aperture creates a slower lens but improves the telecentricity and the depth of field around the operation distance of one focal length $f_1 = 300 \text{ mm}$ in front of the entrance lens. The 2 mm aperture in the StokesCam proved to be a good compromise between image quality and achievable frame rate. With a focal length of $f_2 = 60 \text{ mm}$ of the exit lens, the imaging system has a $\times 1/5$ magnification. One superpixel on the camera, hence, translates to an observed area of around $35 \times 35 \mu\text{m}$.

An exchangeable band-pass filter with a 10 nm width ensures the observation of monochrome light and, therefore, a constant retardance of the wave plate.

1. Data retrieval

In the most simple case, the retrieval of the Stokes vector from the camera intensities consists of the following steps:

1. Dark frame correction: As is common procedure in most camera-based setups, a dark frame is subtracted from the raw data to account for the average

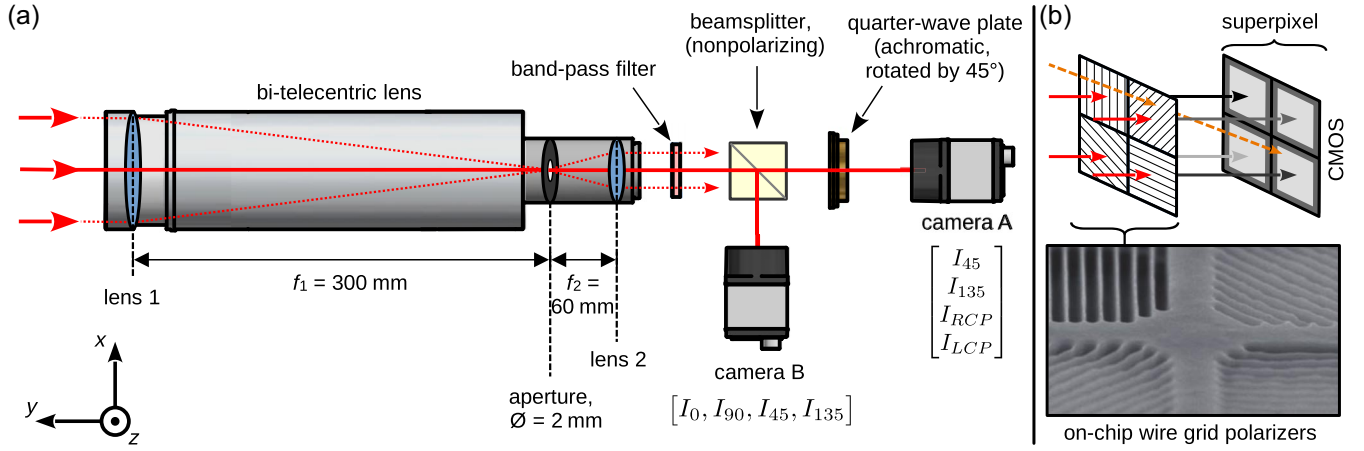


FIG. 2. (a) Setup of the StokesCam. The bi-telecentric lens (not to scale) restricts the light on the entrance and exit side to parallel beams. After passing through an interchangeable band-pass filter with 10 nm FWHM, the light is split into two paths by a nonpolarizing beamsplitter. The MPC B directly measures the linear polarized intensities as shown. The transmitted beam path has a quarter-wave plate with its fast axis rotated to 45° in front of camera A. This would result in the observation of the right and left circular polarized intensities on the 0° and 90° pixels. (b) Schematic of the micropolarizer array in front of the CMOS sensor. A superpixel includes four pixels with the polarizer directions of 0°, 90°, 45°, and 135°. A too-inclined light incidence (dashed line) results in misregistration. Microscopic image by Yamazaki *et al.* [45].

dark current accumulation, defect pixels, and read-out bias of each individual camera. A dark frame should be obtained by averaging several images captured with an integration time matching the exposure time used during operation.

2. Normalization: Because of the different optical paths, inhomogeneous CMOS sensitivity, or even stain, the two camera measurements can not be directly combined into one image. To account for any of those influences, each single pixel must be normalized to its local total intensity S_0 . As in Eq. (13), this is given by the sum of the corresponding orthogonally oriented pixels, e.g., $I'_0 = I_0 / (I_0 + I_{90})$ or $I'_{45} = I_{45} / (I_{45} + I_{135})$. This cross-pairwise normalization has a more favorable error propagation compared with a full superpixel-wise normalization. After this step, S_0 uniformly values one over the full sensor, resulting in the loss of the spatial amplitude information. To prevent this, an image of the superpixel-averaged intensity $S_0 = \frac{1}{4}(I_0 + I_{90} + I_{45} + I_{135})$ of camera A is saved beforehand.
3. Image registration: To resolve the mirroring from the beamsplitter and any small but unavoidable misalignments in the setup, the camera images must be registered (aligned). For this, the image is split into four subimages, according to micropolarizer type (one image containing all 90° pixels, one with 45°, etc.). An affine transformation that preserves parallel lines is then applied to the subimages of camera B to match the position and orientation of camera A. With properly mounted cameras, the transformation matrix needs to be obtained just once. The transform must account for horizontal

mirroring and a small translation and rotation. The image of camera A remains static. A subpixel registration accuracy is achievable with an appropriate target, such as millimeter paper. Although possible [48], images of ordinary objects are not particularly suited for this task. After registration, every pixel of cameras A and B is rescaled with the spatial intensity image of camera A taken in step 2. Since all pixels in the same superpixel, on both cameras, are multiplied by the same factor S_0 , the prior normalization remains intact.

4. Stokes relation: At last, the relations in Eq. (13) are applied superpixel-wise to the preprocessed data to obtain the spatially resolved Stokes parameters. The parameter S_2 is redundant and can be used as an uncertainty estimate. Either both S_2 parameters can be averaged or one of them can be discarded.

This conventional retrieval method assumes accurate angular alignment of the retarder and polarizer axes. It also requires ideal optics with homogeneous and known retardance and polarizer orientations. As shown in the following section, this is not necessarily the case.

2. Component characterization

Given the high accuracy requirements for Mie polarimetry [35,49] (estimated to $\Delta s_i \leq 0.05$), the optical components of the instrument may no longer be considered ideal. It is therefore necessary to characterize the different parts.

The standard procedure to examine a LP P_s uses the Malus law $I = A \cos^2(\alpha + \theta) + I_{\min}$ [see Fig. 3(a)]. A second polarizer P_r is rotated by an angle α in front of P_s . Given the extinction ratio of P_r is high compared with that

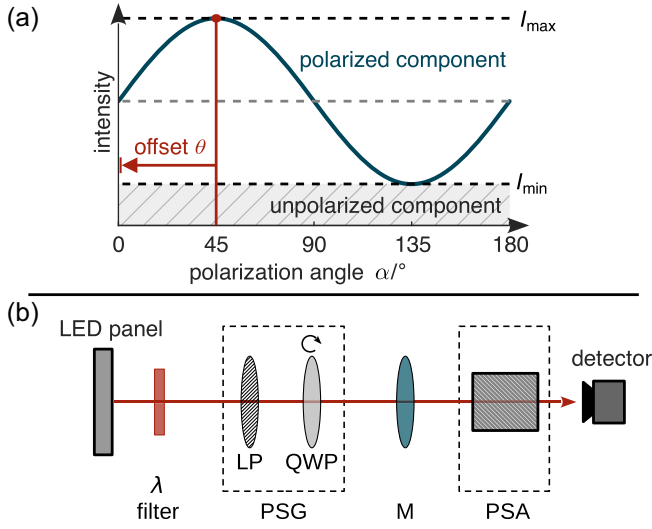


FIG. 3. (a) Intensity curve of the Malus's law $I = A \cos^2(\alpha + \theta) + I_{\min}$. The angular and intensity offsets directly show the angular misalignment and extinction ratio of the tested polarizer. (b) Schematic rundown of the eigenvalue calibration setup that also functions as a Mueller polarimeter. A monochrome light source is followed by the *polarization state generator* (PSG), consisting of a LP and a rotatable QWP. The calibration reference is depicted as M. The *polarization state analyzer* (PSA) converts the incoming polarization into a set of intensities measured by a detector.

of P_s , the intensity passing through both filters is then described by the Malus law. An MPC in place of P_s can directly measure this curve for every pixel. The offset θ to the first maximum then gives the angular orientation of each micro polarizer relative to P_r . The minimal intensity $I_{\min}$ denotes the presence of a component not polarized by the P_s . The quantity $I_{\max}/I_{\min}$ is an estimate for the extinction ratio of P_s .

The result of the Malus analysis for the MPCs is depicted in Fig. 4. Shown are distributions of the micropolarizer's angular misalignment with respect to their intended value. For each camera, the respective mean orientation of all 0° pixels is chosen as the absolute reference. The polarizer directions follow a narrow distribution without visible outliers. The distribution width as well as the average offset goes as far as $\approx 0.2^\circ$, indicating a high precision for such a complex device. Repeated measurements show no sign of temporal or thermal evolution. The latter could be a problem for older types of polarization-sensitive cameras, where the polarizer arrays were glued to the sensor. The measured extinction ratio for the polarizers can be estimated to be at least 100:1, which is consistent with the findings of Nabadda *et al.* [29].

A spatial measurement of the beamsplitter is shown in Fig. 5. A linear horizontal polarized target with $S = (1, 1, 0, 0)^T$ is observed in the reflection direction by camera B, yielding s_1 and s_2 . Ideally, the polarization

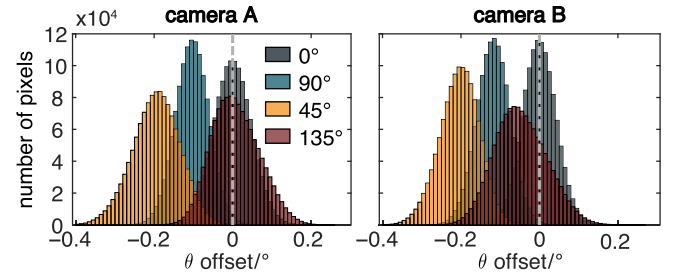


FIG. 4. Angular deviation θ of the micropolarizers from the intended orientation. Distributions are ordered by pixel type and camera. Here, the mean of the 0° pixels on each camera is used as a reference.

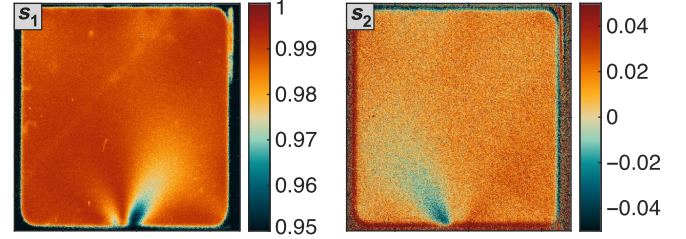


FIG. 5. Measurement of a $S = (1, 1, 0, 0)^T$ target with camera B through the nonpolarizing beamsplitter in reflection. Shown are the linear normalized s_1 and s_2 components. The bottom part shows a clear signal of retardance due to mechanical stress in the glass cube. Notice that the beamsplitter was free of external pressure and not pinch-mounted for this record. The overall deviation from the target in different measurements also suggests the presence of a small but significant birefringence.

should remain unchanged by the nonpolarizing beamsplitter. However, the image shows a slight offset, indicating the presence of a small but significant and unknown birefringence. Also, a strong retardance signal is visible near the bottom, probably induced by mechanical stress in the glass cube originating from the mounting recess. It should be noted that the beamsplitter was free of external pressure and, unlike in standard usage, not mounted during the measurement. The manufacturer, *Thorlabs*, was unable to provide spatial information on the specifications with the required precision, even after further inquiry.

While the quality of polarizers can be readily assessed using Malus's law, (partially) retarding components are more difficult to characterize precisely with an analogous intensity law due to their unfavorable error propagation. Even small influences on the observable intensity curve (e.g., light source stability, angular misalignment, or stain) can lead to major errors in the deduced retardance of a few degrees. Some modern components, like composite wave plates, even require more than one retardance parameter to correctly describe them, further complicating the process. An in-depth characterization of a component's Mueller matrix, on the other hand, requires a previously calibrated

polarimeter of at least similar precision, leading to a chicken-and-egg problem.

This, and the presence of spatial artifacts and inhomogeneities in the optics, concludes that a conventional calibration of the individual polarimeter components is not reasonable at the intended level of accuracy.

B. Eigenvalue calibration

The problem of a virtual reference-free calibration technique, without relying on a priori assumptions about the components, was first solved by Compain *et al.* [30] with the ECM. It has been adapted several times since for different devices and applications [50,51]. Our approach follows the work of Macías-Romero and Török [50], but we show a generalized mathematical framework to accommodate nonsquare matrices. A similar formulation has recently been published by Montes-Gonzalez *et al.* [42]. Combined with small adaptations to the calibration procedure, this enables the application of the ECM to an overdetermined imaging DoA polarimeter.

1. Calibration theory

The underlying setup of the ECM is displayed in Fig. 3(b). Instead of considering every individual component, the polarimeter is treated as a black box, the PSA. The PSA can be described by its device matrix $\mathbf{A} \in \mathbb{R}^{(a \times 4)}$. It translates any incoming (4×1) Stokes vector $\mathbf{S}$ into a number of a intensities, measurable by a detector. In our case, the StokesCam encompasses both the PSA and the detector. With the intensities arranged as a vector $\mathbf{D}$, a polarization measurement translates to

$$\mathbf{D} = \mathbf{A} \cdot \mathbf{S} \leftrightarrow \mathbf{S} = \mathbf{A}^+ \mathbf{D}, \quad (14)$$

where the operation $\mathbf{X}^+$ is the *Moore-Penrose* pseudoinverse of a matrix $\mathbf{X}$. It is the generalization of the matrix inverse $\mathbf{X}^{-1}$ for nonsquare matrices.

For calibration, the setup in Fig. 3(b) additionally includes a monochrome light source, a PSG, and an optical sample $\mathbf{M}$ used as a calibration reference. The PSG is capable of creating a number of g different polarization states (Stokes vectors). This can be designated as the matrix $\mathbf{G} \in \mathbb{R}^{(4 \times g)}$. In our case the PSG consists of a LP and a rotating quarter-wave plate.

Together with the (4×4) Mueller matrix $\mathbf{M}_i$ of the sample in between, the full measurement is represented as

$$\mathbf{D}_i = \mathbf{A} \cdot \mathbf{M}_i \cdot \mathbf{G}, \quad \mathbf{D}_i \in \mathbb{R}^{(a \times g)}. \quad (15)$$

When both $\mathbf{A}$ and $\mathbf{G}$ are known, this equation basically describes a complete Mueller polarimeter that can be used to determine $\mathbf{M}_i$. It is essential that both the PSG and the PSA are Stokes-complete. Thus, the set of generated polarization states must at least include four sufficiently linearly independent states. The PSA, on the other hand,

must be capable of nonambiguously determining every polarization state. Mathematically speaking, this is synonymous with a three-dimensional triangulation on or inside the Poincaré sphere. Unfavorable PSA and PSG designs that generate or sample very self-similar polarization states can substantially impact the achievable precision of a device [52,53].

The ECM solves the problem of the unknown $\mathbf{A}$ and $\mathbf{G}$. For that, the measurement of air as neutral medium is utilized

$$\mathbf{D}_{\text{air}} = \mathbf{A} \mathbf{M}_{\text{air}} \mathbf{G} = \mathbf{A} \mathbf{G}, \quad (16)$$

and its inverse multiplied to Eq. (15), resulting in

$$\begin{aligned} \mathbf{K}_i &= \mathbf{D}_i \cdot \mathbf{D}_{\text{air}}^+, & \mathbf{K}_i &\in \mathbb{R}^{(a \times a)} \\ &= \mathbf{A} \mathbf{M}_i \mathbf{G} (\mathbf{A} \mathbf{G})^+ = \mathbf{A} \mathbf{M}_i \mathbf{A}^+. \end{aligned} \quad (17)$$

Right-side multiplication of $\mathbf{A}$ and rearranging the equation results in the simplified Sylvester equation

$$\mathbf{K}_i \mathbf{A} - \mathbf{A} \mathbf{M}_i = 0, \quad (18)$$

which can be written in a vectorized form as

$$(\mathbf{I}_4 \otimes \mathbf{K}_i - \mathbf{M}_i^T \otimes \mathbf{I}_a) \text{vec}(\mathbf{A}) = 0, \quad (19)$$

$$\mathbf{H}_i \text{vec}(\mathbf{A}) = 0, \quad (20)$$

where $\mathbf{I}_k$ is the $(k \times k)$ identity matrix. Considering a set of calibration samples $\mathbf{M}_1, \mathbf{M}_2, \dots, \mathbf{M}_m$, the Eq. (20) becomes an overdetermined system of equations with $\mathbf{A}$ as the solution. This problem is solvable when converted to a system of normal equations, using a *Gram* matrix $\mathbf{L}$ with

$$\mathbf{L} = \mathbf{H}_1^T \mathbf{H}_1 + \mathbf{H}_2^T \mathbf{H}_2 + \dots, \mathbf{H}_m^T \mathbf{H}_m, \quad (21)$$

which at last gives

$$\mathbf{L} \cdot \text{vec}(\mathbf{A}) = 0. \quad (22)$$

The calibration process can thus be described as a matrix equation and eigenvalue problem. The solution $\mathbf{A}$ is the eigenvector corresponding to the $\lambda = 0$ eigenvalue of the measurable matrix $\mathbf{L}$. The final computation of the eigenvalue spectrum may be done with any suitable algorithm. The matrix $\mathbf{G}$ is accessible with Eq. (16), resulting in a fully calibrated Mueller polarimeter as a side effect.

To ensure a unique solution to Eq. (22) with only one eigenvalue equaling zero, the characteristics of the samples $\mathbf{M}_i$ must be chosen accordingly. The minimal and most convenient choice is a retarder close to a quarter-wave plate, plus a polarizer at 0° and 45° orientations [30].

As of Eq. (18), the Mueller matrix of each calibration sample must be known, so one could argue that the ECM

also needs predefined optical components. However, Eq. (17) shows that $\mathbf{M}_i$ and $\mathbf{K}_i$ are *similar* matrices and share the same eigenvalues. Because $\mathbf{K}_i$ is a measurable quantity, the eigenvalues of the calibration samples are accessible. These can be used to precisely calculate their physical properties. With respect to Eq. (9), many simple polarization optics can be well generalized as a polarizing retarder. It has four eigenvalues (eigenvalue spectrum) with

$$\{r_x^2, r_y^2, (\cos \phi - i \sin \phi) \cdot r_x r_y, (\cos \phi + i \sin \phi) \cdot r_x r_y\}. \quad (23)$$

This model is a very accurate description for LPs (with $\phi \rightarrow 0$) and single crystal or polymer retarders (with $r_x \rightarrow 0, r_y \rightarrow 1$). The Mueller matrices of composite and achromatic wave plates can differ; wherefore, those components should not be utilized as a sample. In the case of $a > 4$, the matrix $\mathbf{K}_i$ yields more than four eigenvalues. The four values that are physically relevant for $\mathbf{M}_i$ are the largest. The agreement of the complex conjugates in Eq. (23) is a good metric for the correctness of the measured eigenvalues associated with retardance. In the rare case of $a \gg g$, it can be convenient to calculate the matrix

$$\mathbf{C}_i = \mathbf{D}_{\text{air}}^+ \cdot \mathbf{D}_i = \mathbf{G} \mathbf{M}_i \mathbf{G}, \quad (24)$$

which is also similar to $\mathbf{M}_i$ and $\mathbf{K}_i$. Both matrices $\mathbf{C}_i$ and $\mathbf{K}_i$ thus have an intersecting set of eigenvalues that are the eigenvalues of $\mathbf{M}_i$.

For the complete identification of the matrices $\mathbf{M}_i$, their exact orientation in the setup is required. Because any Mueller matrix and its rotated counterpart share the same eigenvalues [matrix similarity in Eq. (10)], the angular position cannot be determined by the eigenvalues. Instead, the rotation angle θ_i of each element can be fitted by minimizing the ratio η of the two smallest eigenvalues of $\mathbf{L}$

$$\eta = \sqrt{\frac{\lambda_{2a}}{\lambda_{2a-1}}}. \quad (25)$$

The value η is a valuable metric for the error and convergence of the calibration. Note that all but one sample can be fitted, while one must be defined as the angular reference for the whole polarimeter. An LP with its distinct axes is best in our view.

2. Calibration implementation

For a PSG, we use the setup in Fig. 3(b). A stepper motor rotates the quarter-wave plate to $g = 120$ reproducible positions per circle, with an increment of $(3 \pm 0.06)^\circ$. The reference samples $\mathbf{M}_i$ consist of a wire-grid LP at 0° , 90° , and 45° orientations, as well as a polymer quarter-wave

TABLE I. (a) Parameters of the reference samples, derived from the measured eigenvalues. Values are given as the bootstrapped mean with corresponding 1σ interval. (b) End results and convergence parameters of the ECM. They may serve as a reference for the reader's own implementation. (c) Angular orientations of the reference samples from eigenvalue minimization.

(a) M_{ref} parameters		(b) ECM result	
p_x	0.908 ± 0.001	η	0.106
p_y	0.072 ± 0.020	$\min(\lambda)$	3.6×10^{-3}
r_x	0.993 ± 0.002	κ	2.28
r_y	0.985 ± 0.002		
ϕ	$(-87.8 \pm 0.3)^\circ$		
(c) Reference sample orientations			
θ_r	$(-0.56 \pm 0.25)^\circ$	θ_{p0}	$(-2.6 \pm 0.25)^\circ$
θ_{p90}	$(89.08 \pm 0.25)^\circ$	θ_{p45}	$(0 \pm 0)^{\text{oa}}$

^aThe 45° orientation is used as a reference with zero error per definition.

plate. Since all retarding elements in the PSA, PSG, and the samples are wavelength-dependent, the calibration must be done separately for every intended wavelength.

The eigenvalue calibration of the StokesCam follows a process analogous to Sec. III A 1. Steps 1 to 3 are executed as described. Theoretically the intensity normalization (step 2) is redundant, as the ECM inherently incorporates the different intensity levels of both cameras in $\mathbf{A}$. However, this would restrict the later operation to a fixed ratio of sensor gains and exposure times, which is unfavorable as the main sensitivity for circular polarization exclusively roots in camera A and vice versa. Because the ECM needs the information of S_0 over the different PSG states, the intensity rescaling in step 3 is of importance.

Afterward, the ECM can be done for each superpixel and the intended wavelength as presented, using $a = 6$ pixels of the available 8. The 45° and 135° pixels of camera A have been discarded as redundant. They can be included in the ECM, but we noticed no significant improvement in η convergence from a further overdetermined PSA. The overdetermined PSG with $g = 120$, on the other hand, proved to successfully reduce individual frame noise. We found the presented calibration problem to be numerically stable, resulting in satisfactory convergence despite the application of *Moore-Penrose* pseudoinverses. A decomposition of PSA and PSG into symmetric matrices as proposed by Rosales *et al.* [51] did not prove necessary in our setup. When choosing $g \geq 4$ PSG states for the calibration, a preferably uniform distribution on the Poincaré sphere should be emphasized. An over-representation of certain states can increase the error significantly in the overdetermined calibration. Similar findings were reported by Montes-Gonzalez *et al.* [42]. The presented uncertainties are derived in the next section.

The final convergence result of the center pixel is exemplarily shown in Table I, together with the deduced sample parameters. The resulting $\mathbf{A}^+$ is used to replace step 4 in the data retrieval process (Sec. III A 1). Instead of the relations in Eq. (13), the Stokes parameters are rather calculated from the measured intensities with the device matrix as in Eq. (14). The following section will discuss the potential errors in further detail.

C. Estimation of calibration uncertainty

When discussing the error of a polarimeter, one must distinguish between the systematic and the statistical components. The systematic error is synonymous with the desired error of the calibration. In our case, it originates from limited angular reproducibility of the stepper motor or numerical conditioning during the ECM. The usually far smaller statistical error comes from the image noise of the sensor and can be well reduced by time averaging or superpixel binning. It determines the achievable signal-to-noise ratio (SNR) and the least significant difference in polarization that can be reliably measured [see Eq. (26)], but it does not indicate the accuracy.

An unclear distinction between both error types can lead to some confusion about surprisingly small error bars of measurements [29], especially because the systematic error is difficult to obtain. Another pitfall is validating the device with an allegedly known target, where only air can be considered optically ideal with the identity Mueller matrix of a vacuum [42].

To nonetheless estimate the calibration uncertainty of the StokesCam, a statistical method is necessary. Considering idealized conditions, the converged ECM provides the exact PSA matrix $\mathbf{A}$ and $\mathbf{A}^+$. In reality, $\mathbf{A}$ contains several propagated errors, mainly from the reconstructed parameters of the samples $\mathbf{M}_i$ and their orientations θ , which again are also influenced by the precision of the stepper motor during the calibration process. The latter is around 0.06° in our case. The calibration uncertainty is therefore assessed in a two-step process presented below:

1. Estimating the uncertainty of the derived reference sample parameters via bootstrapping.
2. Calculating the influence of perpetuated reference sample parameters on the ECM result and on a test measurement.

From the total of 120 measured PSG states, 200 subsets of at least 60 states are bootstrapped [54], and the ECM is executed on each set as seen in Sec. III B. Every set that happens to exhibit a low PSG orthogonality, which is identifiable by an unusually high η , should be discarded to avoid statistical outliers. The mean and standard deviation of all resulting sample parameters $p_{x/y}$, $r_{x/y}$, ϕ and their orientations θ are calculated and given in Table I. The largest uncertainties visibly lie in the wave plate retardance and the angular orientations.

Based on the obtained parameter uncertainties, the total error of the PSA is estimated with a Monte Carlo approach, as follows. The ECM is again computed 400 times, including all 120 PSG states. The calibration sample parameters are predetermined by the results of the bootstrapping method. The averages listed in Table I are taken and perturbed by Gaussian noise with one of two possible standard deviations. Half of the calculations (200) use the parameter uncertainty obtained from the bootstrapping as an optimistic noise level (opt). The other half is assigned a more pessimistic noise level of twice this value (pes). This yields two Monte Carlo ensembles of $\mathbf{A}^+$ with different underlying uncertainty scenarios. The undisturbed result $\bar{\mathbf{A}}^+$ from the average parameter values is identical for both ensembles.

Concerning a measurement device, the spread of the reconstructed Stokes parameters is the main interest and also better to visualize than the spread of the individual matrix elements in $\mathbf{A}^+$. For this, the StokesCam measures the polarization signal of a testing setup that consists of a rotating LP with a subsequent (nonideal) quarter-wave plate. The measured Stokes parameters are calculated for all 400 runs and individually compared with the unperturbed results given by $\bar{\mathbf{A}}^+$. The root mean square deviation is then calculated for the optimistic (σ_{opt}) and the pessimistic scenario (σ_{pes}).

In Fig. 6, the Stokes parameters are plotted against the rotation angle α of the polarizer. The reconstruction with $\bar{\mathbf{A}}^+$ as the ECM result is displayed by solid lines (ECM). The Stokes parameters as obtained by the conventional approach (CON) in Eq. (13) without ECM, but using the manufacturer data of the optics, are shown as dashed lines (CON). Major differences up to ± 0.2 are noticeable, especially in s_2 . Naturally, the measured polarization states do not cover the full Poincaré-Sphere but are nonetheless a valuable indicator that the conventional characterization method based on the individual parts is not feasible for high-precision devices.

The gray envelopes in Fig. 6 represent the root mean square deviations from the respective ECM curve for the low (σ_{opt}) and the high noise (σ_{pes}) Monte Carlo ensemble. For the present measurement, this error estimate rests well below 0.04 for the pessimistic uncertainty scenario and throughout below 0.01 for the optimistic error conditions. Notice that these values represent the maxima over the whole range, while the average is even smaller (0.02 and 0.006, respectively). Considering the σ_{pes} as an upper bound, the StokesCam's accuracy is generously estimated to be at least $\Delta s_i \leq 0.04$.

Another relevant parameter for the discussion of numerical errors is the condition number [54] κ of the matrix $\mathbf{A}^+$. It can be seen as a numerical stability estimate of the matrix Eq. (14) $\mathbf{D} = \mathbf{A} \cdot \mathbf{S}$. A small perturbation $\delta \mathbf{D}$ of the intensity measurement $\mathbf{D} + \delta \mathbf{D}$ arises from electrical CMOS noise, Poisson photon noise, or other influences. In

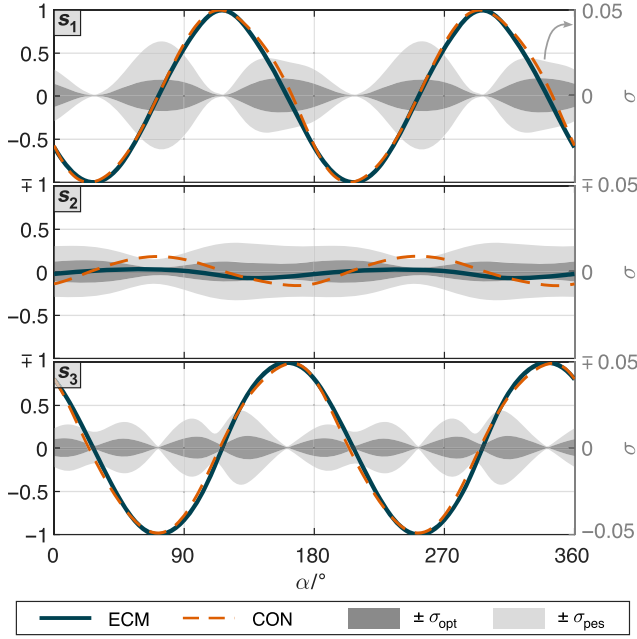


FIG. 6. Normalized Stokes parameters of the test target as measured with the eigenvalue calibration (ECM, solid line) and the conventional method (CON, dashed line). The parameter's root mean square deviations from the Monte Carlo ensembles are depicted as gray envelopes, corresponding to the right axis. The corresponding low (σ_{opt}) and high perturbation (σ_{pes}) scenarios of the reference sample parameters are shown.

this case, the relation for the induced error level in S is given by

$$\frac{\|\delta S\|}{\|S\|} \leq \kappa(\mathbf{A}) \frac{\|\delta D\|}{\|D\|}, \quad (26)$$

with $\|\cdot\|$ being the 2-norm. In our case, we find $\kappa = 2.28$, so that the relative variability of the Stokes parameters is approximately double the relative perturbation of the registered intensities.

The amplitude of the combined dark noise and readout noise for the employed CMOS sensor type lies in the order of 2ADU for 250 ms exposure time at room temperature. Considering the high dynamic range of 12 bit = 4096 ADU, the photon shot noise is the main contributor to the total intensity variance. The reference measurements in Fig. 6 provide a laboratory environment for evaluating the noise amplitude of the resulting Stokes parameters. We observed a root mean square deviation of ≤ 0.005 over the sensor area, depending on total intensity. This translates to an average SNR of 90 dB with a range of 20 to 160 dB. The polarization angles Ψ and Δ exhibit a reduced SNR of 80 dB due to the unfavorable error propagation of Eq. (7).

In conclusion, this results in an estimated accuracy of ≤ 0.04 and a precision of ≤ 0.005 for the StokesCam. It is comparable to a DoT imaging polarimeter by Shibata *et al.* [48].

Further improvements on the architecture of imaging polarimeters to reduce the achievable SNR have been discussed by Tyo [24]. Those, however, encompass polarization setups not in conformance with the available 0° , 90° , 45° , and 135° micropolarizer sensors. Reducing the calibration uncertainty would mainly involve an even more precise PSA with better step precision.

IV. MIE POLARIMETRY OF NANOPARTICLE CLOUDS

Now that the necessary calibration procedure and the potential precision of the StokesCam have been discussed, the polarimeter is deployed for imaging Mie-polarimetry of nanodusty plasmas.

Other literature introducing alternative imaging polarimeter builds often shows stress polarization of polymers [42] or commercial polarization targets [26,29] as exemplary images. These images impressively show the qualitative capabilities of polarimetry in applications such as industrial process and quality control. However, the major challenge of high-precision full-Stokes polarimetry in various fields is the precise measurement of even weak circular polarized light. This counts especially for laboratory operations working with scattering and radiation transport models.

Light scattered by nanoparticles confined in a low-pressure plasma is a complex but fitting example for the high demand of accuracy on polarimetric devices. The nanoparticles are synthesized in the plasma from a reactive gas component and grow from molecular precursors to several hundred nanometers in size. Polarimetry can be used to determine the *in situ* time-dependent particle radius. The method relies on a fitting scattering model and precise data. The circular part of the polarization is of particular importance for this and must be correctly measured even for near-zero values and under low-intensity conditions.

In the following, we therewith present a first glance at the StokesCam's capabilities and show the future potential of the device for the characterization of nanoparticle synthesis in such reactive plasmas.

The particles produced in the synthesis process [31] are highly spherical. For this, the analytical Mie-theory can be applied as the appropriate scattering model for those particles [55]. The measurements shown are directly connected to the work of Greiner *et al.* [31], Groth *et al.* [27], and Petersen *et al.* [56]. As this work primarily concentrates on the hardware side of the polarimetry, a detailed treatise on the implementation of the scattering model and the inverse calculation of the particle properties can be found in Groth *et al.* [38] and Schmitz *et al.* [49].

A. Experimental setup

The vacuum chamber and electrical design for the plasma generation are identical to the setup used in

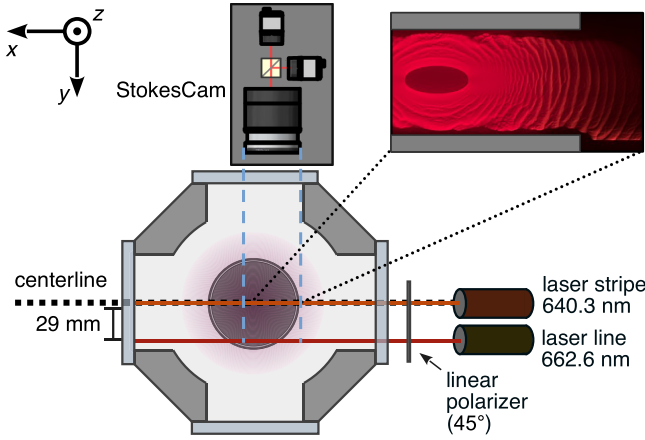


FIG. 7. Top view of the experimental setup for polarimetry measurements on nanodust clouds. The rf-driven plasma is centered above the lower electrode. Two different lasers illuminate the particles. The 640.3 nm laser stripe passes through the center of the cloud. A 662.6 nm laser line is parallelly displaced by 29 cm. The StokesCam measures the scattered light under 90° . The top right image shows a view of the nanodust cloud between the two electrodes illuminated by the red laser stripe. Further details on the plasma chamber are found in Schmitz *et al.* [49].

Schmitz *et al.* [49]. The capacitively coupled rf-discharge is driven at 13.56 MHz with argon as the working gas and a constant gas flow of 8 sccm. Acetylene (C_2H_2) can be added as a reactive component to initiate the nanoparticle synthesis. The amorphous carbon-hydrate particles (a-C:H) continuously grow in a multistage process [36] and exhibit a cylinder-symmetrical distribution in the electrode gap [57].

Two lasers are used as shown in Fig. 7. One 2D laser stripe with a 640.3 nm wavelength that passed through the center of the dust cloud. A laser beam with 662.6 nm is parallelly displaced by 29 cm. Both lasers are linearly polarized at $+45^\circ$. This geometry is similar to Petersen *et al.* [56] and Kobus *et al.* [58] and allows a comparison with their work. The StokesCam is mounted perpendicular to the laser path behind a window access, observing one half of the nanodust cloud under a 90° scattering angle.

Two measurement cycles of the nanoparticle synthesis are conducted. Each cycle starts with a pure argon discharge at 20 Pa pressure, to which a 20% acetylene admixture is added at time $t = 0$ s. The presented imaging polarimetry is conducted during each particle growth cycle with a frame rate of 0.5 Hz. One cycle is measuring the scattering of the 640.3 nm stripe and the other the scattering of the 662.6 nm laser line. The PSA calibration has to be determined for both wavelengths. Also, the suitable wavelength filters are mounted in the StokesCam, respectively.

B. Single beam measurement

We first discuss the single beam measurement of the 640.3 nm laser line. Pixel binning over a small area of the

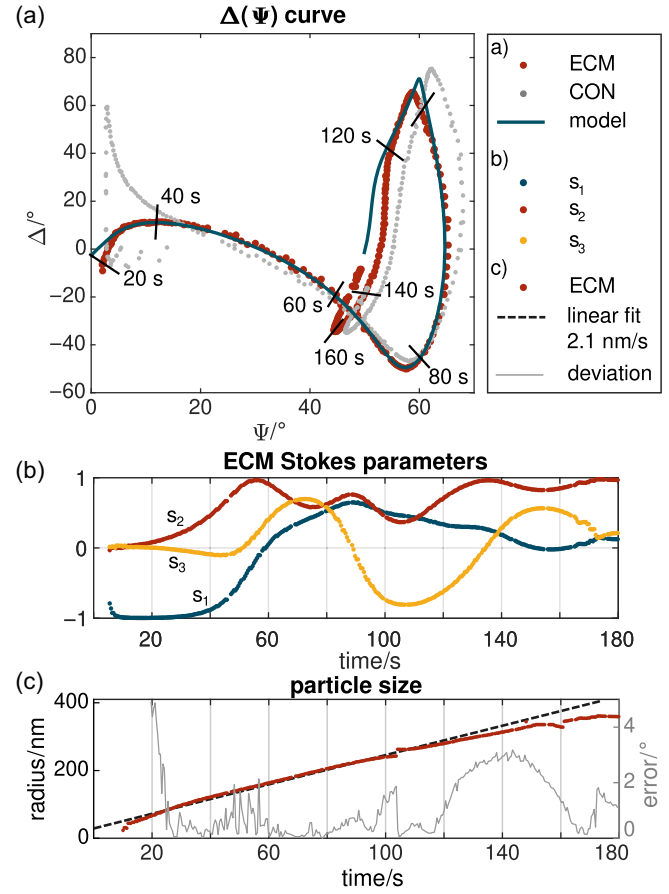


FIG. 8. (a) Polarization of the scattered laser beam light over time in $\Delta(\Psi)$ representation. The area of intensity binning is indicated in red in Fig. 9. Shown are the data points as obtained using the eigenvalue calibration (ECM) and the conventional method (CON). The fitted scattering model with a refractive index of $n = 1.79 + 0.02i$ is given as a solid line. (b) ECM polarization curve in Stokes representation. (c) Radius of the nanoparticles, calculated from the measured $\Delta(\Psi)$ curve (ECM) and fitted refractive index. A linear fit in the interval $t = [10, 120]$ s yields an average growth rate of 2.1 nm/s. The Pythagorean distance of each $\Delta(\Psi)$ data point to the model curve is given as a measure of error.

beam center [red mark in Fig. 9] is applied to increase the SNR. The resultant polarization curve is shown in Fig. 8 in Stokes and $\Delta(\Psi)$ representation [see also Eq. (7)]. The latter is a common visualization for polarization in ellipsometry and dusty plasmas [31]. The measured data is displayed twice: once as obtained with the ECM and once with the standard method without the calibration (CON). Similar to the error assessment in Fig. 6, both methods lead to distinct results. The strongest deviation arises for particular small (radius $a \leq 100$ nm) and large particles $a \geq 220$ nm. In this case, forgoing the ECM would necessarily lead to significant errors in those regimes. In the intermediate part (40 s to 80 s), both methods roughly coincide.

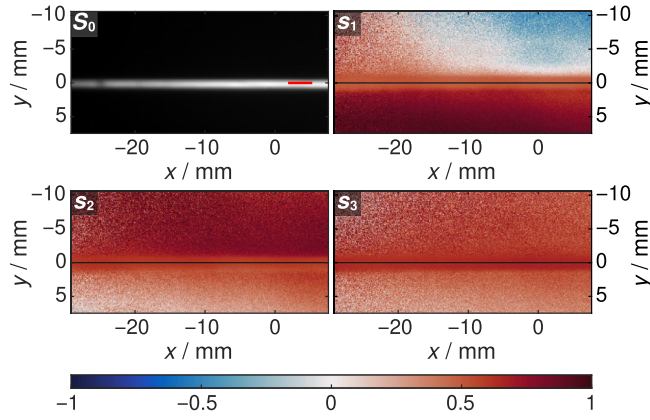


FIG. 9. Spatial polarization signal of the 662.6 nm laser line at 77 s and 200 nm particle radius. Stokes parameters s_1 to s_3 are normalized to the polarized intensity. The vertical position of the laser line at $y = 0$ mm is marked. The asymmetry of multi-scattering effects is clearly visible around this line. The red mark in the image of the total intensity S_0 indicates the averaging area for Fig. 8.

Using the CRAS-Mie method [38], a scattering model is fitted onto the ECM polarization curve to obtain the particle refractive index $n = 1.79 + 0.02i$. Other free parameters are the scattering angle and laser polarization,

where small but significant deviances to the target values are found in the order of $\mathcal{O}(0.1^\circ)$. The model curve is depicted with a solid line and agrees very well with the ECM measurement, even at larger particle sizes. The influence of optical depth, multiscattering events, and a broadening of the size distribution on polarization signals has been discussed in other works [35,59].

The uncalibrated CON values could not be properly evaluated with our model, especially with the visible deviation of Δ at times ≤ 40 s. This is a good indicator that the StokesCam performs well under low-intensity conditions and provides the accurate linear and especially circular polarization.

After determining the refractive index of the $\Delta(\Psi)$ -curve with our scattering model, the respective particle radii can be calculated for data points (see Schmitz *et al.* [49] for details). The angular distance of the data points to the model curve gives a measure of error. A comparison to a linear fit in the time interval of $t = [10 \text{ s}, 120 \text{ s}]$ reveals an almost constant growth rate of 2.1 nm/s, which is in agreement with the findings of Petersen *et al.* [56]. The jump discontinuities at 100 and 160 s arise from a numerical artifact when the model curve and measurement slightly disagree at extremes [e.g., see Fig. 8(a) at $t = 100$ s].

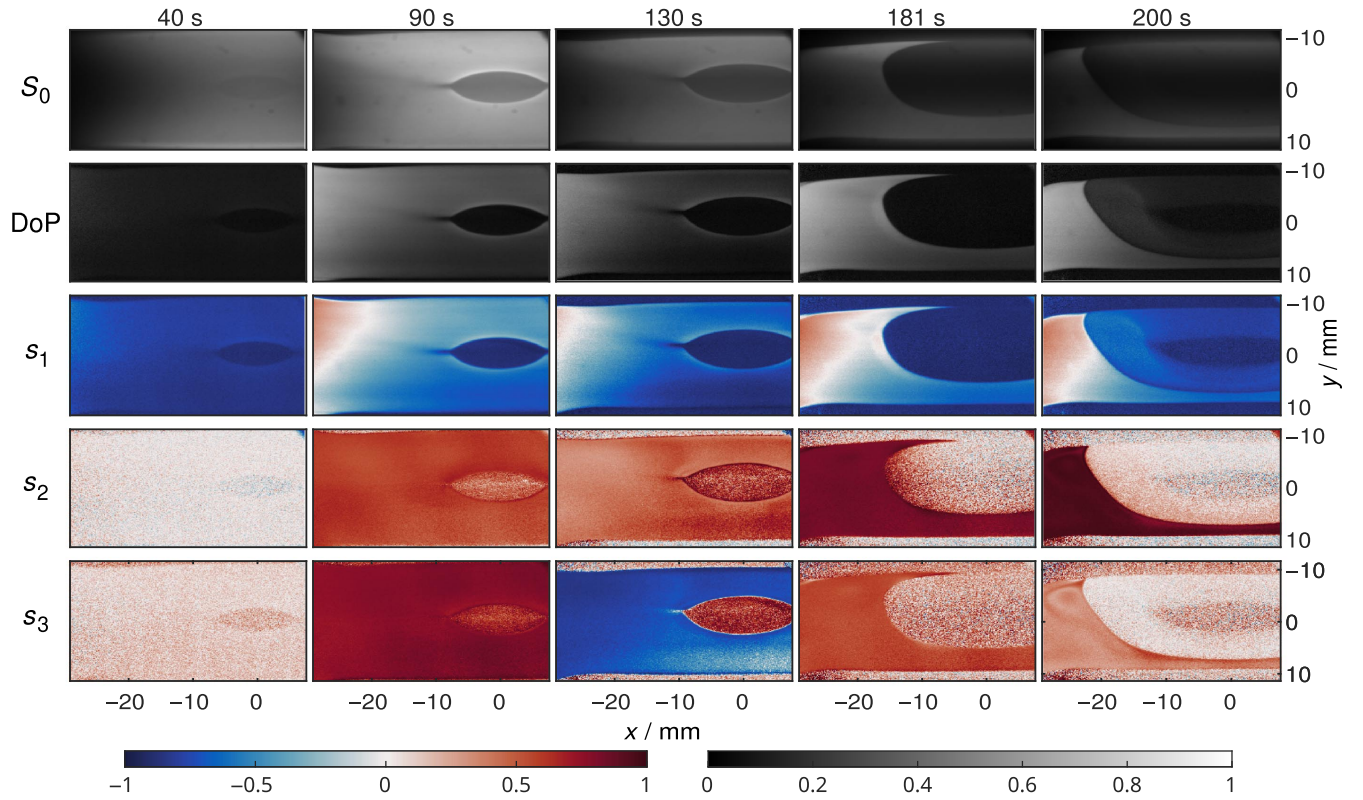


FIG. 10. Spatially resolved polarization of light scattered by the nanoparticle cloud at different times. Stokes parameters s_1 to s_3 are normalized to the polarized intensity, while S_0 is the total intensity is seen by an ordinary camera. The DoP is defined as in Eq. (4). All images are 4×4 binned on the superpixels. Note that the vertical scale is not congruent with Fig. 9.

Another significant subject of investigation is the influence of multiscattering events on Mie polarimetry and other light-scattering techniques. Recent efforts in the modeling of those effects on nanodusty plasmas [35,60] have been made, but a precise experimental validation is lacking [58]. With the StokesCam, this is now within the scope of future work. A first example of the multiscattering events observed with the 662.6 nm laser line is presented in Fig. 9 for a particle radius of 200 nm. The vertical position of the beam is indicated as a solid black line. Above and below the laser beam, distinct polarization patterns arising from the scattering asymmetry [58] are visible. The S_0 image, as would be seen by standard nonpolarization-sensitive cameras, does not register any perceivable multiscattering. This underlines the sensitivity of Stokes polarimetry to the presence of nanoparticles in the volume.

C. Laser stripe measurement

The measurement of the laser stripe traversing the centerline of the cloud is presented in Fig. 10. Displayed is the scattered polarization as normalized Stokes parameters as well as the total intensity at different representative time steps. Similar to Fig. 9, the nanoparticles are significantly clearer detectable in the Stokes polarization signal than by the total intensity S_0 of the scattered light alone. Since Stokes parameters are based upon normalized intensity differences, the influence of stain on the optics is also greatly reduced. Both features make imaging polarimetry particularly suitable for computer vision. Overall, this will have a significant impact on the perspective-monitoring capabilities of nanoparticle synthesis in reactive or processing plasmas.

Not only can the presence of the particles be well detected but also the distribution and evolution of the cloud over time. The changing particle size generates a strong signal in the Stokes components s_1, s_2, s_3 , as well as the spatially calculable DoP. The expansion of the central, particle-free void [61] and the later formation of new particle generation in the center (*layered growth* [27]) are clearly visible and more pronounced than in S_0 .

Combining the measured polarization with a suitable light scattering model for the particles, as in Fig. 8, can yield the spatially resolved particle radius and optical properties. Such a model, though, should encompass the presence of multiscattering and nonmonodisperse size distributions, especially for the denser areas of the cloud [58].

V. CONCLUSION AND OUTLOOK

We presented the StokesCam, a high-resolution full-Stokes polarimeter based on two MPCs. The rigid DoA and focal plane setup operates without moving or variable components and is therefore particularly suitable for

low-vibration or nontemperature-controlled environments. A detailed introduction to the measuring principle and necessary polarimeter architecture was given, using the Stokes-Mueller formalism. The utilized MPCs have been characterized concerning their polarization angle and precision. Furthermore, we demonstrated that commercially available optics can exhibit significant unwanted and uncharacterized birefringence, leading to substantial systematic errors in a polarimeter. To address the problem of calibrating such devices without a credible reference or a priori assumptions, we implemented a variant of the ECM by Compain *et al.* [30]. We generalized the calibration method to overdetermined imaging polarimeters and showed that the application of pseudo matrix inverses does not introduce further numerical instabilities for the presented setup.

Using a comprehensive error estimation, we analyzed the potential systematic and statistical errors of the calibration method. We used a bootstrapping and Monte Carlo approach instead of relying on validation measurements of allegedly known targets. The upper bound for the systematic uncertainty was found to be $\Delta s_i \leq 0.04$, with comparatively negligible statistical noise from the sensor. We stretch so that the spatial and temporal detector noise is not an appropriate measure of the polarimeter precision. The minimal signal resolution lies at $\delta s_i = 0.002$. The uncertainties could potentially be reduced further with a more precise stepper motor.

As a first glance into the perspective of imaging nanoparticle diagnostics, we presented Mie polarimetry measurements of nanodusty plasmas. The polarization signal possesses a clear advantage over simple intensity data regarding SNR and the tracking of the particle size. We have shown that a suitable light-scattering model can be used to robustly infer the particle's time-dependent radius and optical properties. A significant aspect for the modeling of imaging polarization data in the presence of multiscattering events is the scattering asymmetry we managed to show.

The StokesCam thus enables further investigations of nanoparticle synthesis in reactive plasmas, such as the study of multiscattering events, particle size distributions, and spatial growth dynamics.

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DATA AVAILABILITY

All data supporting the findings of this article are openly available from Schmitz [62].

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