Abstract

Polarization dynamics gives insight on the changes in the polarization state of partially polarized beams. We present a measurement system to follow the femtosecond scale polarization fluctuations from a point of a random light field that is based on scattering scanning near-field microscopy. To ensure the method will provide details on the instantaneous properties of an incident beam, we construct equations for the electric field scattered from a subwavelength, dipolar nanoparticle. The polarization dynamics of the scattered far field prove that apertureless SNOM can be used to follow the rapid variations of the polarization ellipse as long as the nanoprobe has a response time that is in the femtosecond range. We establish basic conditions for the polarizability and shape of the nanoparticle to guarantee that the dipole can react to the variations in the incident beam on the same timescale. The criteria indicates that the selected nanoparticles’ response times are within 2-37 femtoseconds. The most suitable nanoparticles for use with our method are silver Archimedean solids ranging from 92-136 nm in size. A limitation for this measurement approach is that no information on the instantaneous circular polarization states can be determined.

Keywords: Polarization; Femtosecond phenomena; Near-field microscopy; Scattering particles; Nanomaterials.
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One vital characteristic of light is its polarization. The polarization properties describe the behavior of the electric field components of an optical wave. The polarization properties give information on the orientation of the electric field and the phase difference between the electric field’s orthogonal components also [1–4]. Knowing the polarization characteristics of light is important for subjects such as imaging [5], light and matter interactions [6, 7], and designing optical elements and/or devices to tailor the electromagnetic field [8]. With the recent scientific breakthroughs from nanotechnology, these same areas are also crucial for studying nanomaterials [9, 10] and developing nanoscale optical devices [11–13]. In addition to the topics mentioned above, another key subject of interest is how the polarization of light can change during propagation and how these polarization changes can be studied.

Typically the polarization measurements of fields are averages; however, the state of polarization can fluctuate for random fields. Fluctuations in the polarization state are due to the intrinsic properties of light sources and media where the light propagates. Details on the correlations between polarization and the electric field generated from the inherent qualities of light and matter are covered in statistical optics [3, 4, 14–16]. The possibility of applications for pulse shaping [17], quantum information [18], optical sensing [19], and telecommunications [20] has led to the expansion of research on spectral variations to the instantaneous polarization properties of vector beams.

For vectorial light, most of the conceptual and experimental work on fluctuating fields has been proposed for the spatial domain. Some examples of the theoretical formulations for the spatial propagation of random beams include the study of electro-
magnetic beams modulated by random phase screens [21], changes in the polarization of partially coherent beams that travel in dielectric media [22] and free space [23], fluctuations in the instantaneous Stokes parameters for quasi-monochromatic electromagnetic beams [24], and the probability density functions of the instantaneous Stokes parameters for scattered light [25]. The synthesis of a fluctuating beam for the analysis of its statistical properties [26] and the design of a polarimetric interferometer that detects the spatial polarization fluctuations of polarized speckle fields [27] has been experimentally demonstrated recently also.

When considering the principles dictating the temporal properties of polarization dynamics, the main theory has been described for partially polarized electromagnetic beams in terms of the instantaneous Jones vector [28] and the instantaneous Stokes vector [29, 30]. There are also additional theoretical studies on polarization variations for pulses traveling through time lenses [31] and the instantaneous polarization ellipse of few-cycle pulsed beams [32]. The amount of experimental work in the time domain has been limited because of the difficulties in detecting the rapid variations in random beams since these changes occur within femtoseconds (detectors currently have a temporal resolution in the picosecond range) [33].

A common method for examining instantaneous polarization properties has been modulating light to be detected within microseconds or nanoseconds so that the constraints posed by detectors are not present [34]. So far, these techniques are mostly suited for monochromatic beams. The measurements do not include the polarization dynamics properties given in Ref. [29] such as the polarization time and polarization correlation function. The only study that provides measurements for general beams (any type of photon statistics and any spectral range) and their polarization properties governed by polarization dynamics has been presented in [35]. This technique employs a modified Michelson interferometer with a detection scheme that is sensitive to two-photon absorption to determine femtosecond scale polarization fluctuations. However, this method does not allow for the determination of the instantaneous polarization states and one is not able to determine the source of the polarization fluctuations.

Here we introduce a new measurement scheme to observe temporal polarization dynamics with apertureless near-field scanning microscopy (SNOM). We consider random light, which is two-dimensional (beam-like) and stationary, and its interaction with a nanoparticle that has an induced dipole moment. The electric field
at the site of the dipole and far from the probe are calculated using Maxwell’s
equations and retarded potentials. After obtaining the electric far field, we derive
formulas for the polarization matrix, instantaneous Stokes parameters, and polar-
ization correlation functions of the far field. The polarization correlation function
and the properties of convolution for polarization in the time domain show that it
is possible to measure polarization variations as long as the nanoprobe can scatter
light fast enough to follow the changes in the polarization state of the incident beam.
Therefore, we use the time domain’s representation of the dipole moment and polar-
izabilities of isotropic nanoparticles to estimate the response times of nanoparticles.
All of the nanoparticles’ response times are within the femtosecond range, which
further proves the potential of this method. A limitation of our method is that
the orientation of the electric field and observation direction does not allow one to
examine the instantaneous left- and right-handed circularly polarized states. This
problem may be solved by developing new electric far field equations with different
observation directions.

This work is divided into six parts. We begin by reviewing the essential prop-
erties of random light in Chapter 2. Chapter 3 provides the theory for polarization
fluctuations in the time domain. Next, we discuss techniques for measuring the
polarization properties of light and whether these methods are useful for investigat-
ing fluctuating fields in Chapter 4. Chapter 5 illustrates the principles of detect-
ing instantaneous polarization properties with apertureless SNOM and the relevant
equations needed for measurements. The criteria for choosing nanoparticles to act
as probes is explained in Chapter 6. Finally, a summary of this work is included in
the Conclusions.
Chapter II

Basic Concepts of Random Light

Natural, or chaotic light, stems from the process of spontaneous emission. The light generated consists of multiple electromagnetic waves that propagate in multiple directions and have random phases. Given that natural light originates from a random process, we may characterize the random light using statistics [3, 4, 14–16]. This chapter introduces the main correlation functions of the electromagnetic field for random light. We primarily focus on the temporal coherence and temporal polarization properties of random light fields.

2.1 Description of the electromagnetic field

We begin by defining the random light as a stationary, fluctuating electromagnetic beam propagating along the \( z \)-axis. For a given point \( \mathbf{r} \) in space at time \( t \), the electric vector of the field is expressed by the complex analytic signal \( \mathbf{E}(\mathbf{r}, t) = [E_x(\mathbf{r}, t), E_y(\mathbf{r}, t)]^T \), where \( T \), \( E_x \), and \( E_y \) denote the transpose, the \( x \)-component of the electric field vector, and the \( y \)-component of the electric field vector. The coherence properties of the light at times \( t \) and \( t + \tau \) for points \( \mathbf{r}_1 \) and \( \mathbf{r}_2 \) are given by the second-order \( 2 \times 2 \) electric mutual coherence matrix [3, 36]

\[
\mathbf{\Gamma}(\mathbf{r}_1, \mathbf{r}_2, \tau) = \langle \mathbf{E}^*(\mathbf{r}_1, t) \mathbf{E}^T(\mathbf{r}_2, t + \tau) \rangle. \tag{2.1}
\]

Here the brackets and asterisk indicate the ensemble average and the complex conjugate, respectively. The second-order mutual coherence matrix is quasi-Hermitian and is also expressed as [29]

\[
\mathbf{\Gamma}(\mathbf{r}_1, \mathbf{r}_2, \tau) = \mathbf{\Gamma}^\dagger(\mathbf{r}_1, \mathbf{r}_2, -\tau), \tag{2.2}
\]
where the dagger represents the Hermitian adjoint.

The mutual coherence matrix can be utilized for describing the degree of coherence. For vectorial fields, the degree of coherence (in squared form) is given by [3,36]

$$\gamma_{EM}^2(r_1, r_2, \tau) = \frac{\text{tr}[\Gamma(r_1, r_2, \tau)\Gamma(r_2, r_1, -\tau)]}{\text{tr}[\Gamma(r_1, r_1, 0)]\text{tr}[\Gamma(r_2, r_2, 0)]},$$  \hspace{1cm} (2.3)

where \(\text{tr}\) represents the trace, or sum of the diagonal elements, of a matrix. The subscript EM denotes an electromagnetic field. The electromagnetic degree of coherence provides insight on the variation of vectorial light’s intensity and polarization state in terms of the Stokes parameters (discussed later) [29,36,37]. The squared degree of electromagnetic coherence is determined by using Young’s interferometer to measure the sums of the visibility of fringes that are created due to the interference of Stokes parameters. The degree of electromagnetic coherence ranges from zero to one. When the electromagnetic degree of coherence is equal to zero, there are no changes in the polarization or intensity of light in interference. As the degree of coherence approaches the upper limit, the intensity of light and/or the Stokes parameters are modulated [37].

2.2 Polarization properties of the field

The polarization of an electromagnetic beam is dependent on the relative complex amplitude(s) of the component(s) of the electric field. Thus, the polarization of light follows the oscillations in the electric field over time. As the electric vector of deterministic light oscillates with time, the polarization vector draws an ellipse that establishes the state of polarization. The polarization ellipse can simplify to a line or a circle. When the polarization ellipse becomes a line, light is considered to be linearly polarized. The plane of polarization can be along or in between the axes of the electric vector components. For a polarization ellipse that becomes circular, the electric field is thought to be circularly polarized. Depending on the orientation of the oscillations, light can be left- or right-circularly polarized light [1–4]. This behavior of the polarization ellipse is expected for deterministic light. However, the polarization ellipse of a random beam varies over time and can be an ellipse, line, or circle [14–16,29,38]. Information on the average polarization state can be evaluated
using the following quantities: polarization matrices, Stokes parameters, and the degree of polarization.

### 2.2.1 Polarization matrices

The polarization properties of the random field at a single point can be extracted from the polarization matrix, which is given by the second-order mutual coherence matrix of Eq. (2.1) at a single time and point [3, 29]:

\[
J(r) = \Gamma(r, r, 0)
\]

\[
= \begin{bmatrix}
J_{xx}(r) & J_{xy}(r) \\
J_{yx}(r) & J_{yy}(r)
\end{bmatrix},
\]

where \(J_{ij}(r) = \langle E^*_i(r, t) E^T_j(r, t) \rangle\), with \((i, j) = (x, y)\). Diagonal elements \(J_{xx}(r)\) and \(J_{yy}(r)\) represent the average intensities in the \(x\)- and \(y\)-direction. The off-diagonal elements \(J_{xy}(r)\) and \(J_{yx}(r)\) express the correlations between the \(x\)- and \(y\)-component of the electric field. The diagonal elements of the polarization matrix are real values and the off-diagonal elements are complex [3]. The polarization matrix is purely Hermitian [29].

### 2.2.2 Stokes parameters

The polarization of an electromagnetic beam can be further expressed with the Stokes parameters. The Stokes parameters are [3, 36]

\[
S_0(r) = J_{xx}(r) + J_{yy}(r),
\]

\[
S_1(r) = J_{xx}(r) - J_{yy}(r),
\]

\[
S_2(r) = J_{yx}(r) + J_{xy}(r),
\]

\[
S_3(r) = i[J_{yx}(r) - J_{xy}(r)].
\]

\(S_0(r)\) is the trace of the polarization matrix and gives the average intensity of the electromagnetic field. The polarization state of the beam is defined with the other Stokes parameters. \(S_1(r)\) is the intensity difference of the \(x\)- and \(y\)-component of the electric field, which describes linearly polarized light along the \(x\)- and \(y\)-axis. Light polarized 45° between the \(x\)- and \(y\)-axis is provided by \(S_2(r)\). The last Stokes parameter, \(S_3(r)\), illustrates the intensity difference of left-handed and right-handed circularly polarized light. All of the Stokes parameters are averages and real quantities [36].
2.2.3 Degree of polarization

To understand how much of the electromagnetic beam is polarized, we introduce the degree of polarization (DOP). The degree of polarization specifies the ratio of the intensity of polarized light to the intensity of the total field. The DOP can be mathematically written as [3]

\[ P_2(r) = \sqrt{1 - \frac{4 \det J(r)}{\text{tr}^2 J(r)}}, \quad (2.9) \]

where \( \det \) is the determinant and the subscript 2 designates that this degree of polarization is for a two-dimensional field. The DOP is constrained to the following range of values [3]:

\[ 0 \leq P_2(r) \leq 1. \quad (2.10) \]

Light with a degree of polarization equal to zero is considered as unpolarized light, which means that the light has no preferred polarization state (completely random polarization). A degree of polarization with the value of one occurs when light has a permanent polarization state, or is fully polarized. Values between zero and one imply that an electromagnetic beam is partially polarized. Partially polarized light has a preferred polarization state, which is represented by the polarized part of the field [3].

Alternatively, the degree of polarization can be expressed with the Stokes parameters [37]:

\[ P_2^2(r) = \sum_{n=1}^{3} s_n^2(r), \quad (2.11) \]

where \( s_n(r) \) represents the normalized Stokes parameters and is given as

\[ s_n(r) = \frac{S_n(r)}{S_0(r)}, \quad n = 0, \ldots, 3. \quad (2.12) \]

The normalized Stokes parameters can be any value from \(-1 \leq s_n(r) \leq 1\). Using the Stokes parameters to define the degree of polarization provides information on the changes in the polarization state when a beam is allowed to interfere with itself [37]. This representation of the degree of polarization resembles the electromagnetic degree of coherence since the sums of the visibility of fringes for a beam interfering with itself within a Young’s interferometer is equivalent to the normalized Stokes parameters [37].
We previously introduced the polarization properties of random beams. These properties of light are only true for time averages of the electromagnetic field over long periods of time. When examining light over much shorter timescales, one gets insight on the fluctuations of the electromagnetic field [29]. This chapter describes the polarization variations, or polarization dynamics, of fluctuating light.

3.1 Definition

Fully polarized light and monochromatic light do not undergo any changes in the polarization state or polarization ellipse. However, the intensity can change for fully polarized light. When considering partially polarized light, the polarization ellipse experiences several fluctuations in its shape and orientation within timescales typically on the order of femtoseconds. We define these temporal variations as polarization dynamics. Although polarization dynamics occurs for artificial and natural light sources, during very small periods of time, one can find segments along fully unpolarized beams where the polarization state is essentially unchanged. The interval of time that the polarization state of partially polarized beams remains unaltered is identified as the polarization time $\tau_P$ [29,30]. The polarization dynamics of any two partially polarized fields, with the same time-averaged state of polarization and degree of polarization, are not necessarily similar [30,33,35].
3.2 Mathematical interpretation of polarization dynamics

To follow the fluctuations in the polarization ellipse, we must consider the instantaneous polarization properties of the electromagnetic field. We present the instantaneous Stokes parameters as [35]

\begin{align*}
S_0(t) &= I_x(t) + I_y(t), \\
S_1(t) &= I_x(t) - I_y(t), \\
S_2(t) &= I_{+45}(t) - I_{-45}(t), \\
S_3(t) &= I_{RCP}(t) - I_{LCP}(t).
\end{align*}

The subscripts \(x\), \(y\), \(+45\), and \(-45\) correspond to the intensities of light linearly polarized along the \(x\)-axis, \(y\)-axis, at an angle of \(+45^\circ\), and at an angle of \(-45^\circ\). \(I_{RCP}\) and \(I_{LCP}\) describe the intensities of right- and left-circularly polarized light [35]. The instantaneous Stokes parameters have real values since they are intensity differences. The instantaneous Stokes parameters satisfy the relation [29]

\[
S_0(t) = \sqrt{S_1^2(t) + S_2^2(t) + S_3^2(t)},
\]

which demonstrates that a source of light has a distinct polarization state that evolves with time. The single-point Stokes parameters can be determined by taking the time average of the instantaneous Stokes parameters over an unbounded time span [29].

3.2.1 Poincaré sphere

We can describe the instantaneous polarization state with the Poincaré sphere. The Poincaré sphere graphically illustrates the changes in the instantaneous Stokes parameters over a time interval of \(t\) to \(t + \tau\) (see Fig. 3.1). The fluctuations in the polarization state are shown with the assistance of the Poincaré vector [29]

\[
\mathbf{S}(t) = \hat{s}(t)S_0(t),
\]

where \(\hat{s}(t)\) is a unit vector parallel to the Poincaré vector. The unit vector can be expressed with the instantaneous Stokes parameters [29]:

\[
\hat{s}(t) = \left[ \frac{S_1(t)}{S_0(t)}, \frac{S_2(t)}{S_0(t)}, \frac{S_3(t)}{S_0(t)} \right].
\]
Given the relationship between the instantaneous Stokes parameters in Eq. (3.5), the magnitude of the Poincaré vector is equal to one, which is also the radius of the Poincaré sphere [29].

![Figure 3.1: The Poincaré sphere. The unit vectors \( \hat{s}(t) \) and \( \hat{s}(t + \tau) \), which are separated by an angle \( \theta \), define the polarization state at times \( t \) and \( t + \tau \). The red line shows the path of \( \hat{s}(t) \) over the time interval \( \tau \).]

The Poincaré vector specifies a point on the surface of the Poincaré sphere that is a linear combination of the instantaneous Stokes parameters. When the polarization state changes, the Poincaré vector will trace a path along the surface of the Poincaré sphere. When the polarization state does not change, the Poincaré vector will not move, but the intensity \( S_0(t) \) will vary. If \( \langle S(t) / S_0(t) \rangle = 0 \), the point will lie at the origin and is considered to represent unpolarized light. Light is partially polarized when the time averaged Poincaré vector lies inside of the Poincaré sphere. For fully polarized light, the Poincaré vector is fixed [29].

### 3.2.2 Polarization correlation function

The dynamics of a polarization state can also be analyzed with weighted intensity correlations. The similarities in the Poincaré vectors at time \( t \) and \( t + \tau \) are described by the fourth-order polarization correlation function [29]

\[
\gamma_p(\tau) = \frac{\langle S(t) \cdot S(t + \tau) \rangle}{\langle S_0(t) S_0(t + \tau) \rangle}.
\] (3.8)
Here the brackets indicate a time average. The time average provides a comparison of the instantaneous polarization states over multiple cycles of time length \( \tau \). Since the numerator of the polarization correlation function can be simplified to \( \langle |\mathbf{s}(t) \cdot \mathbf{s}(t + \tau)| \rangle S_0(t)S_0(t + \tau) \), it is obvious that

\[-1 \leq \gamma_P(\tau) \leq 1. \tag{3.9}\]

The limits of Eq. (3.9) represent the orthogonal and the same Poincaré vector. We also see that \( \gamma_P(\tau) = 1 \) when \( \tau = 0 \), which indicates that the light at a time \( t \) has a specific polarization state. It is important to note that the two polarization states become more similar as the value of \( \gamma_P(\tau) \) increases [29].

### 3.2.3 Polarization time and polarization length

The polarization time can be determined using the polarization correlation function. The polarization time is identified as the time where \( \gamma_P \) decreases to a specific value, which is arbitrary. Figure 3.2 illustrates the polarization correlation of fully polarized light, partially polarized light, and unpolarized light.

![Figure 3.2](image-url)

**Figure 3.2:** Illustration of the polarization correlation function as a function of \( \tau \) for beams with different degrees of polarization, \( P_i \) (i=1, 2, 3 and 4). The uppermost curve corresponds to \( P_1 \), which is for a fully polarized beam. The second curve corresponds to \( P_2 \), which is for a partially polarized beam with a degree of polarization greater than 1/2. The third curve corresponds to \( P_3 \), which is for a partially polarized beam with a degree of polarization less than 1/2. The last curve corresponds to \( P_4 \), which is unpolarized light. \( \tau_{P_i} \) are the polarization times for each beam [29].
We see that $\tau_P$ can be finite or infinite. The polarization time is only a finite value when the beam is unpolarized or is a partially polarized beam with a degree of polarization less than 0.5. Therefore, $\gamma_P(\tau)$ should correspond to a value specifically for these types of beams. We choose to define the polarization time as the time $\gamma_P(\tau) = 0.5$. Moreover, the fact that $\tau_P$ for unpolarized light is finite shows that unpolarized light can appear to exhibit polarization dynamics when the time interval is very small [29].

We introduce the polarization length $l_P$, which is defined as the average length where the polarization state of an electromagnetic wave is not significantly altered. The polarization length is expressed as [29]

$$l_P = c \tau_P,$$  \hspace{1cm} (3.10)

where $c$ is the speed of light in a vacuum.
We have introduced the time-averaged polarization properties in Chapter II and the instantaneous polarization properties in Chapter III. The focus of this chapter is to describe some techniques for characterizing the polarization of electromagnetic fields: interferometry, polarimetry, and scanning near-field optical microscopy (SNOM). We consider if these approaches can be useful for characterizing polarization fluctuations.

4.1 Interferometry

The use of interference to obtain information on correlations for electromagnetic beams is known as interferometry. The interference of beams usually occurs by superimposing two beams that have propagated different optical path lengths. Interference creates spatial intensity fringes that can be studied with interferometers [1,3,4,37]. Common interferometers include Michelson’s interferometer, Young’s interferometer, the Mach-Zehnder interferometer, and the Hanbury Brown-Twiss intensity interferometer. The Michelson interferometer studies temporal changes in interference by adjusting the optical system to detect light that has traveled two different optical path lengths, which arrive at the detector at different times. Young’s interferometer studies spatial interference patterns, and a Mach-Zehnder interferometer can study both spatial and temporal interference [1,3,4]. See Fig. 4.1 for the optical systems of the Young and Michelson interferometer.

Regarding polarization, interferometers can be employed to primarily study the degree of polarization for optical beams. The degree of polarization has been experimentally determined by measuring the intensities and visibilities of the four Stokes parameters with a Young’s interferometer [37]. Another experiment has investigated
Figure 4.1: Young’s interferometer (a). A planar light source incident on two slits propagates as a spherical wavefront that experiences constructive and deconstructive interference. These interferences can be seen as fringes at the detector. Michelson’s interferometer (b). The general Michelson interferometer consists of a beam splitter (BS), two mirrors (M1 and M2), and a detector. Changing the distance of one of the mirrors from the beam splitter induces intensity and polarization state variations at the detector.

the degree of polarization as a function of phase modulations and the second Stokes parameters ($S_2$) with a Mach-Zehnder interferometer [39]. Theoretically, a Hanbury Brown-Twiss interferometer can be used to obtain the degree of polarization by measuring the intensity correlations [36].

It is possible to exploit these methods of interferometry to study the fluctuations of partially polarized light when the fields are beam-like and obey Gaussian statistics. For Gaussian light, the instantaneous polarization properties can be studied by using second-order correlations instead of fourth-order correlations, which can be measured with interferometers [33,36]. When considering general two-dimensional beams, instantaneous polarization properties can only be attained with detectors that have ultrafast response times. Currently, the fastest detectors have response
times in the picosecond range, which is too slow to measure polarization dynamics [33]. Another option for examining polarization fluctuations of general beams is to modify an interferometer’s optical setup to be sensitive to ultrafast photon processes that can be detected.

Only one experiment, to our knowledge, has been able to measure the instantaneous polarization properties of beams in the time domain [35]. The intensity correlations of light sources (general and Gaussian) were measured using a Michelson interferometer that utilizes 2-photon absorption. This method has allowed for the determination of the correlations of the instantaneous Stokes parameters, $\gamma_P(\tau)$ given by Eq. (3.8), polarization time $\tau_P$, and polarization length $l_P$ given by Eq. (3.10). However, this approach does not directly measure the properties of the instantaneous polarization state or provide information on what optical components or processes are affecting the changes in the polarization.

4.2 Polarimetry

Polarimetry is a group of measurement techniques that determine the polarization properties of an electromagnetic field that interacts with an optically active medium or a series of optical elements such as polarizers and waveplates [1]. When the incident light interacts with the medium, the light can be reflected, scattered, or refracted towards a detector, which gives information about the electromagnetic field at the site of the medium [6]. When the electromagnetic waves interact with polarizing components, the light that is detected has undergone a transformation that can be described with matrices [4]. There are many types of polarimeters, but Stokes polarimeters are currently the main devices used to study the polarization of electromagnetic waves [9]. Two recent examples of Stokes polarimeters are based on the spin-orbit interaction (SOI) of light [40] and plasmonics [41].

4.2.1 Spin-orbit based polarimeter

The Stokes polarimeter exploiting the SOI of light consists of a subwavelength scatterer that interacts with multiple optical modes (see Fig. 4.1) [40]. The spin of the incident beams determines which direction the scattered light will propagate since each mode has a different response, which is due to the quantum spin Hall effect. The power measurements of each mode is connected to the incident field and effec-
tive area of the nanoscatterer. Measuring the power of each waveguide allows the polarization matrix to be calculated. Once the polarization matrix has been calibrated, the measured and calibrated polarization matrix can be used to determine the state of polarization in terms of the Stokes parameters [40].

![Stokes polarimeter based on spin orbit interaction. A 400 × 220 nm T-shaped nanostructure scatters incident light differently depending on the polarization state of light and whether the incident light is perpendicular or parallel to the scatter. Optical modes interacting with the polarimeter are indicated with blue, red, green, and yellow. The power measurements corresponding to each polarization provide insight on the Stokes parameters of the incident light. Taken from Ref. [40].](image)

**Figure 4.2:** Stokes polarimeter based on spin orbit interaction. A 400 × 220 nm T-shaped nanostructure scatters incident light differently depending on the polarization state of light and whether the incident light is perpendicular or parallel to the scatter. Optical modes interacting with the polarimeter are indicated with blue, red, green, and yellow. The power measurements corresponding to each polarization provide insight on the Stokes parameters of the incident light. Taken from Ref. [40].

### 4.2.2 Plasmonic polarimeter

The plasmonic Stokes polarimeter establishes the state of polarization and Stokes parameters of a light source by scanning the incident field over linear and spiral slits formed in a gold thin film that has been fabricated above a silicon based Schottky-detector [41]. There are six slits in total, and a pair of slits correspond to each of the four Stokes parameters. The incident field will cause maximum absorption inside a slit when the opening of the slit and the polarization of the electric field are orthogonal to one another. The absorption of each slit is measured using a photodetector. Evaluating the photocounts as a function of the degree of linear
polarization, or as the phase difference between the $x$- and $y$-component of the incident beam, gives insight on the polarization state [41]. An illustration of this polarimeter is provided in Fig. 4.3.

Figure 4.3: Nanopolarimeter based on plasmonics. Focusing a laser over each slit in the gold thin film allows for direct measurement of the absorption of the light by the silicon detection setup. The absorption values correspond to the Stokes parameters of the incident laser. Taken from Ref. [41].

4.2.3 Using polarimeters for following polarization fluctuations

Although the plasmonic and SOI Stokes polarimeters provide novel methods for discovering the polarization state, the techniques are spatial. Therefore, polarization dynamics cannot be studied with these devices. It is only possible to examine the instantaneous polarization properties of light with polarimeters that allow temporal measurements and the optically active medium directs the incident field toward the detector within femtoseconds. Thus, polarimeters that measure the response of an incident field that has interacted with a medium may be utilized for observing polarization fluctuations. While there are no known polarimetric devices that can analyze polarization dynamics, one polarimeter has been able to successfully verify a fluctuating field’s degree of polarization and the Stokes parameters as a function of the angular orientation of linearly polarized light [42].
4.3 Scanning near-field optical microscopy

Scanning near-field optical microscopy (SNOM) has revolutionized microscopes by enhancing the systems’ spatial resolution into the nanometer range beyond the classical diffraction limit. Scanning near-field optical microscopes have different working configurations, but a key method of SNOM is scattering SNOM (apertureless scanning near-field optical microscopy). Apertureless SNOM detects light in the far field that has been scattered by a subwavelength probe that scans the near field. The detection of the electromagnetic field far from the site of the nanoprobe gives insight on the properties of the incident beam at the probe [43].

Scattering scanning near field optical microscopy is typically used for measuring the intensity of light in the near field, but some studies have focused on obtaining polarization properties of polarized fields. A recent work has managed to obtain the polarization matrix of scattered partially polarized beams with a waveplate and polarizer [44,45]. This method is spatial and occurs in the frequency domain. However, the same procedure can be performed in the time domain and could provide information on the instantaneous field properties.

In comparison to interferometry, apertureless SNOM and polarimetry offer measurements of the polarization properties of beams regardless of the statistics of the light. When considering scattering SNOM and polarimetry, scanning near-field optical microscopy is the better approach for following temporal polarization fluctuations. Polarimeters typically do not measure the far field: therefore, polarimeters cannot follow the polarization dynamics of an incident field. Other advantages of scattering SNOM include its ability to investigate polarization fluctuations for near fields and the spatial resolution of the technique in comparison to polarimetry.
Chapter II and Chapter III have described the key factors of fluctuations that occur in random light. Chapter IV has highlighted several methods for analyzing polarization properties of light and has explained how nanoparticles could be employed for measuring the instantaneous polarization state. All of these chapters together provide the complete framework necessary for developing a new approach to experimentally witness polarization dynamics. This chapter puts forward a different means for investigating fluctuations in the polarization state.

5.1 Proposed measurement technique

We start by considering a fluctuating, stationary electromagnetic beam traveling in the z-direction. The total electric field is represented by the electric vector \( \mathbf{E}(r, t) = [E_x(r, t), E_y(r, t), 0]^T \). The polarization matrix gives the average local polarization state and degree of polarization of the incident field. Now a polarizable nanoparticle probe that is much smaller than the wavelength of the incident light is inserted into the electromagnetic field. The nanoprobe can be treated as a point-like electric dipole with a dipole moment \( \mathbf{P} \) proportional to the local electromagnetic field as [2]

\[
\mathbf{P}(r_0, t) = \int_{-\infty}^{t} \chi_e(t - t') \mathbf{E}(r_0, t') dt',
\]

(5.1)

where \( r_0 \) is the position of the nanoparticle and \( \chi_e \) is the 3×3 electric susceptibility tensor of the nanoparticle. The light scattered by the electric dipole is observed from the far field with femtosecond scale time resolution. As mentioned in the previous chapter, the properties of light in the far field are dependent on the local electric
field properties at the site of the nanoparticle. Therefore, it is possible to study polarization properties for any point along an electromagnetic wave.

5.2 Deriving the electric far field

We develop the electromagnetic field beginning from the macroscopic Maxwell’s equations (in Gaussian units) [2]

\[
\nabla \times \mathbf{B} - \frac{1}{c} \frac{\partial}{\partial t} \mathbf{E} = -\frac{4\pi}{c} \mathbf{j}, \tag{5.2}
\]

\[
\nabla \times \mathbf{E} - \frac{1}{c} \frac{\partial}{\partial t} \mathbf{B} = 0, \tag{5.3}
\]

\[
\nabla \cdot \mathbf{E} = -4\pi \rho, \tag{5.4}
\]

\[
\nabla \cdot \mathbf{B} = 0, \tag{5.5}
\]

where \( \mathbf{B} \) is the magnetic induction, \( \mathbf{E} \) is the electric field strength, \( c \) is the speed of light, \( \mathbf{j} \) is the free current density, and \( \rho \) is the free charge density. The free current density and the free charge density can further be expressed as [2]

\[
\mathbf{j} = \frac{\partial}{\partial t} \mathbf{P} + c (\nabla \times \mathbf{M}), \tag{5.6}
\]

\[
\rho = -(\nabla \cdot \mathbf{P}). \tag{5.7}
\]

Here \( \mathbf{P} \) and \( \mathbf{M} \) are the polarization and magnetization. The polarization describes the polarization density due to electric dipoles, and the magnetization describes the magnetic density due to magnetic dipoles.

We now define \( \mathbf{B} \) and \( \mathbf{E} \) in terms of an arbitrary vector \( \mathbf{A} \) and an arbitrary scalar function \( \phi \). \( \mathbf{A} \) and \( \phi \) are chosen so that \( \mathbf{E} \) and \( \mathbf{B} \) satisfy Maxwell’s second and fourth equations [2]:

\[
\mathbf{B} = \nabla \times \mathbf{A}, \tag{5.8}
\]

\[
\mathbf{E} = -\frac{1}{c} \frac{\partial}{\partial t} \mathbf{A} - \nabla \phi. \tag{5.9}
\]

With these definitions of the electric field strength and magnetic induction, Maxwell’s first and third equations are written as

\[
\nabla^2 \mathbf{A} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{A} - \nabla (\nabla \cdot \mathbf{A} + \frac{1}{c} \frac{\partial}{\partial t} \phi) = -\frac{4\pi}{c} \mathbf{j}, \tag{5.10}
\]

\[
\nabla^2 \phi - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \phi + \frac{1}{c} \frac{\partial}{\partial t} (\nabla \cdot \mathbf{A} + \frac{1}{c} \frac{\partial}{\partial t} \phi) = -4\pi \rho. \tag{5.11}
\]
When considering the Lorentz condition [2]

\[ \nabla \cdot A + \frac{1}{c} \frac{\partial}{\partial t} \phi = 0, \]  

Eq. (5.10) and Eq. (5.11) become the inhomogeneous wave equations [2]

\[ \nabla^2 A - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} A = -\frac{4\pi}{c} j, \]  

\[ \nabla^2 \phi - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \phi = -4\pi \rho. \]  

The Lorentz condition is consistent with the continuity condition [2]

\[ \frac{\partial}{\partial t} \rho + \nabla \cdot j = 0. \]  

Instead of using \( A \) and \( \phi \), we can introduce the Hertz vectors \( \Pi_e \) and \( \Pi_m \), which are also known as the electric polarization potential and the magnetic polarization potential. \( A \) and \( \phi \) are expressed in terms of the Hertz vectors as [2]

\[ A = \frac{1}{c} \frac{\partial}{\partial t} \Pi_e + \nabla \times \Pi_m, \]  

\[ \phi = -(\nabla \cdot \Pi_e). \]  

The Hertz vectors have the same mathematical relation to \( A \) and \( \phi \) in Eq. (5.16) and Eq. (5.17) as \( j \) and \( \rho \) to \( P \) and \( M \) in Eq. (5.6) and Eq. (5.7). Using these relations, the inhomogeneous wave equations [Eq. (5.13) and Eq. (5.14)] become [2]

\[ \nabla^2 \Pi_e - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \Pi_e = -4\pi P, \]  

\[ \nabla^2 \Pi_m - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \Pi_m = -4\pi M. \]  

From this point we will only consider the electric field. After substituting Eq. (5.16) into Eq. (5.9), the electric field can be presented as [2]

\[ \mathbf{E} = -\frac{1}{c} \frac{\partial}{\partial t} \left( \frac{1}{c} \frac{\partial}{\partial t} \Pi_e + \nabla \times \Pi_m \right) + \nabla (\nabla \cdot \Pi_e). \]
Using $\nabla \nabla \cdot \equiv \nabla \times \nabla \times + \nabla^2$ and the condition
\[ \nabla^2 \Pi_e - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \Pi_e = 0, \] (5.21)
Eq. (5.20) becomes [2]
\[ E = \nabla \times \left( \frac{1}{c} \frac{\partial}{\partial t} \Pi_m + \nabla \times \Pi_e \right). \] (5.22)

To consider the electric field scattered by the electric dipole, we assume a dipole situated at $r_0$, the site of the probe, that has its orientation characterized by unit vector $\hat{n}$. Such an electric dipole moment $\mathbf{P}$ can be described by the following polarization [2]:
\[ \mathbf{P}(r, t) = p(t) \delta(r - r_0) \hat{n}(t), \] (5.23)
where $p$ is the electric polarization as a function of time and $\delta$ is the Dirac delta function. In our case, the unit vector is allowed to change instantaneously and is thought to be within the $xy$-plane. Since $\hat{n}$ is not fixed, the dipole moment simplifies to
\[ \mathbf{P}(r, t) = p(t) \delta(r - r_0). \] (5.24)

The general equation for the electric Hertz vector is [2]
\[ \Pi_e = \int \frac{[\mathbf{P}]}{R} \, dV', \] (5.25)
where the brackets indicate that the polarization is described as a retarded potential, $R$ is the magnitude of the distance between $r$ and $r'$, and $dV'$ is the differential volume of the entire space. When substituting the polarization for the electric dipole moment [given by Eq. (5.24)] into Eq. (5.25), the Hertz vector becomes
\[ \Pi_e = \int \frac{p(t - R/c) \delta(r' - r_0)}{R} \, dV \] (5.26)
\[ = \frac{p(t - R/c)}{R} \] \[ = \frac{[\mathbf{P}]}{R}, \]
where $R$ is the magnitude of the distance between $\mathbf{r}$ and $\mathbf{r}_0$.

Since the magnetization vector is zero, $\mathbf{\Pi}_m = 0$ holds everywhere in space and Eq. (5.22) can be rewritten as

$$E = \nabla \times (\nabla \times \mathbf{\Pi}_e).$$

(5.27)

Using the identities $\nabla \times \nabla \times \equiv \nabla \nabla \cdot - \nabla^2$ and $\nabla^2 \mathbf{\Pi}_e = (1/c^2)\ddot{\mathbf{\Pi}}_e$, Eq. (5.27) simplifies to [2]

$$E = \nabla (\nabla \cdot \mathbf{\Pi}_e) - \frac{1}{c^2} \ddot{\mathbf{\Pi}}_e.$$

(5.28)

Here $\ddot{\mathbf{\Pi}}_e$ means to take the partial derivative of the variable with respect to time twice.

With the knowledge of Eq. (5.26) and that there is no $z$-component of the polarization vector, the scattered electric field can be simplified to the following equation (see Appendix for full derivation):

$$E = \left\{ \frac{3[p_x(t)]}{\sqrt{2}R^4} + \frac{3[\dot{p}_x(t)]}{\sqrt{2}cR^3} + \frac{[\ddot{p}_x(t)]}{\sqrt{2}c^2R^2} \right\} \mathbf{R} - \left\{ \frac{[\mathbf{p}(t)]}{R^3} + \frac{[\dot{\mathbf{p}}(t)]}{cR^2} + \frac{[\ddot{\mathbf{p}}(t)]}{c^2R} \right\}. $$

(5.29)

Here $\dot{p}_x$ means to take the partial derivative of the variable with respect to time and $\mathbf{R}$ is a vector that denotes the observation direction, which is fixed. $\mathbf{R}$ is given by the following equation:

$$\mathbf{R} = R\hat{\mathbf{u}}_R,$$

(5.30)

where $\hat{\mathbf{u}}_R$ is the radial unit vector of the spherical polar coordinate system. The other two unit vectors are $\hat{\mathbf{u}}_\theta$ and $\hat{\mathbf{u}}_\varphi$. The unit vectors are given by

$$\hat{\mathbf{u}}_R = \sin\theta\cos\varphi \hat{\mathbf{u}}_x + \sin\theta\sin\varphi \hat{\mathbf{u}}_y + \cos\theta \hat{\mathbf{u}}_z,$$

(5.31)

$$\hat{\mathbf{u}}_\theta = \cos\theta\cos\varphi \hat{\mathbf{u}}_x + \cos\theta\sin\varphi \hat{\mathbf{u}}_y - \sin\theta \hat{\mathbf{u}}_z,$$

(5.32)

$$\hat{\mathbf{u}}_\varphi = -\sin\varphi \hat{\mathbf{u}}_x + \cos\varphi \hat{\mathbf{u}}_y.$$

(5.33)

Here $\hat{\mathbf{u}}_x$, $\hat{\mathbf{u}}_y$, and $\hat{\mathbf{u}}_z$ are the Cartesian coordinate system unit vectors. The angles $\theta$ and $\varphi$ specifying the observation direction are considered to be $45^\circ$ and $0^\circ$, which simplifies the unit vectors to

$$\hat{\mathbf{u}}_R = \frac{1}{\sqrt{2}}(\hat{\mathbf{u}}_x + \hat{\mathbf{u}}_z),$$

(5.34)

$$\hat{\mathbf{u}}_\theta = \frac{1}{\sqrt{2}}(\hat{\mathbf{u}}_x - \hat{\mathbf{u}}_z),$$

(5.35)

$$\hat{\mathbf{u}}_\varphi = \hat{\mathbf{u}}_y.$$
A visual representation of the electric field scattered from an electric dipole can be seen in Fig. 5.1.

Figure 5.1: Spherical polar coordinate system for an electric dipole. The observation direction is indicated by the vector $\mathbf{R}$, which is the distance of a point along an electromagnetic wave from the site of the probe (origin), and the vector $\mathbf{P}$ is the dipole moment. $\hat{u}_R$, $\hat{u}_\theta$, $\hat{u}_\phi$ are the unit vectors of the spherical polar coordinate system.

The electric far field can be determined by eliminating the higher order exponential terms in the denominator of Eq. (5.29) and expressing the second half of the equation in terms of the unit vectors of the spherical polar coordinate system:

$$E_{\text{far}} = -\left\{ \frac{[\dot{p}_x(t)]}{\sqrt{2}c^2 R} \hat{u}_\theta + \frac{\sqrt{2}[\dot{p}_y(t)]}{\sqrt{2}c^2 R} \hat{u}_\phi \right\}. \quad (5.37)$$
5.3 Polarization properties of the scattered electric field

At this point we can determine the polarization properties of the far field. By substituting Eq. (5.37) into Eq. (3.1)–(3.4), we get the instantaneous Stokes parameters:

\[ S_0(t) = \left| \dddot{p}_x(t) \right|^2 + 2 \left| \dddot{p}_y(t) \right|^2 \]
\[ S_1(t) = \frac{\left| \dddot{p}_x(t) \right|^2 - 2 \left| \dddot{p}_y(t) \right|^2}{2c^4R^2}, \]
\[ S_2(t) = \frac{2\sqrt{2}\left| \dddot{p}_x(t) \right| \left| \dddot{p}_y(t) \right|}{2c^4R^2}, \]
\[ S_3(t) = 0. \]

Given that \( S_3(t) \) is zero, there is no right-handed or left-handed circularly polarized light observed in the far field. This is consistent with the fact that dipole radiation is linearly polarized.

Using the equation for the scattered electric far field and Eq. (2.4), the polarization matrix becomes

\[ \mathbf{J}_{\text{far}}(\mathbf{r}) = \frac{1}{2c^4R^2} \begin{pmatrix} \langle |\dddot{p}_x(t)|^2 \rangle & \langle \sqrt{2} |\dddot{p}_x(t)| |\dddot{p}_y(t) \rangle \rangle \\ \langle \sqrt{2} |\dddot{p}_y(t) \rangle |\dddot{p}_x(t) \rangle \rangle & \langle 2|\dddot{p}_y(t)|^2 \rangle \end{pmatrix}. \]

Now we can calculate the polarization correlation function by inserting the instantaneous Stokes parameters given by Eq. (5.38)–(5.41) into Eq. (3.8):

\[ \gamma_P(\tau) = \frac{\langle (|\dddot{p}_x(t)|^2 - 2|\dddot{p}_y(t)|^2)(|\dddot{p}_x(t + \tau)|^2 - 2|\dddot{p}_y(t + \tau)|^2) \rangle}{\langle (|\dddot{p}_x(t)|^2 + 2|\dddot{p}_y(t)|^2)(|\dddot{p}_x(t + \tau)|^2 + 2|\dddot{p}_y(t + \tau)|^2) \rangle} + \frac{\langle (2\sqrt{2}|\dddot{p}_x(t)| |\dddot{p}_y(t) \rangle) (2\sqrt{2}|\dddot{p}_x(t + \tau)| |\dddot{p}_y(t + \tau) \rangle) \rangle}{\langle (|\dddot{p}_x(t)|^2 + 2|\dddot{p}_y(t)|^2)(|\dddot{p}_x(t + \tau)|^2 + 2|\dddot{p}_y(t + \tau)|^2) \rangle}. \]

If the instantaneous polarization state does not vary from \( \tau = 0 \) to \( \tau = \tau_P \), then \( \gamma_P(\tau) \) reduces to one, which means that the light has a specific polarization state. Also, the instantaneous Poincaré vectors \( \mathbf{s}(t) \) and \( \mathbf{s}(t + \tau) \) are equal. Moreover, the polarization time is the half the width of the FWHM of the polarization correlation function’s peak.
Suitable Nanoparticles for Scattering Light

In the last chapter we have shown that it is possible to measure instantaneous changes in the polarization state. The suggested method is only possible if the electric dipole can scatter light to the far field within femtoseconds. This chapter establishes the criteria for selecting appropriate nanoparticles to act as probes.

6.1 Basic assumptions of the nanoparticles

We first consider the nanoparticle to be isotropic, or to have uniformity in all orientations. Given the definition of isotropy, the shape, the material, and the electric polarizability matrix $\alpha(\omega)$ of the nanoparticle has restraints. For a nanoparticle to be isotropic, the shape is usually a sphere or any of the geometries defined by the cubic symmetry point group (cube, tetrahedron, rhombicuboctahedron, etc), and the nanoparticle is usually composed of glass or metal [46]. We consider the electric polarizability matrix of the form [47]

$$\alpha(\omega) = \begin{pmatrix} \alpha_{xx} & 0 & 0 \\ 0 & \alpha_{yy} & 0 \\ 0 & 0 & \alpha_{zz} \end{pmatrix},$$  \quad (6.1)$$

where $\alpha_{xx}$, $\alpha_{yy}$, and $\alpha_{zz}$ describe the response of the particle to an electric field applied in the $x$-, $y$-, and $z$-direction. For $\alpha(\omega)$ to be isotropic, all of the diagonal elements must be equal. This means that the polarizability matrix is [47]:

$$\alpha(\omega) = \alpha(\omega)I,$$  \quad (6.2)$$
where $\alpha(\omega)$ and $I$ are the polarizability and the $3\times3$ identity matrix.

Since polarizabilities are given as a function of wavelength in Refs. [48–51], we must next formulate a connection between polarizabilities as a function of wavelength, as a function of angular frequencies, and as a function of time. We first relate the central wavelength $\lambda_0$ and the bandwidth $\sigma_\lambda$ at the FWHM of the peak to the central angular frequency $\omega_0$ and the bandwidth in angular frequencies $\sigma_\omega$. The central wavelength can be converted into the central angular frequency by [1]

$$\omega_0 = \frac{2\pi c}{\lambda_0}. \quad (6.3)$$

The bandwidth in angular frequencies is determined by [6]

$$\sigma_\omega = \frac{2\pi c}{\lambda_0^2} \sigma_\lambda. \quad (6.4)$$

The electric susceptibility in the time domain is related to the polarizability in the frequency domain by Fourier transform:

$$\chi_e(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \alpha(\omega) \exp(i\omega t) d\omega. \quad (6.5)$$

We consider the response of the polarizability in the frequency domain to be Gaussian, which simplifies the polarizability in the time domain to

$$\chi_e(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \exp \left[ -\frac{(\omega - \omega_0)^2}{2\sigma_\omega} \right] \exp(i\omega t) d\omega$$

$$= \frac{\sigma_\omega}{2\pi} \sqrt{\frac{\pi}{2}} \exp(i\omega_0 t) \exp \left( -\frac{t^2}{2\sigma_\omega^2} \right) I. \quad (6.6)$$

This makes the response in the time domain Gaussian as well. The quantity $1/\sigma_\omega$ defines the bandwidth in the time domain $\sigma_t$. The electric susceptibility in terms of $\sigma_t$ can be expressed as

$$\chi_e(t) = \frac{\sqrt{2\pi}}{2\sigma_t \pi} \exp(i\omega_0 t) \exp \left( -\frac{t^2}{2\sigma_t^2} \right) I. \quad (6.7)$$

### 6.2 Assumptions for the light’s polarization

The dipole moment introduced in Chapter V can be considered as a time-invariant and causal system [52]. With the properties of this system, we can describe the
relationship between the functions given in Eq. (5.1). The electric susceptibility is
given as response function and does not depend on the time at which the nanoparticle
will be inserted into an electromagnetic field. Since the system is causal, the dipole
moment of the nanoparticle is not dependent on times larger than \( t \). We also see
that the polarization of the far field is related to the electric field at the site of the
probe due to the time-translation invariance of the electric susceptibility. Given
these relationships, we consider nanoparticles with electric susceptibility response
times in the femtosecond range so that the polarization dynamics can be analyzed
at the far field. We can estimate the response time of the electric susceptibility with
Eq. (6.6) by determining the bandwidth at the FWHM for different nanoparticles.
The response time corresponds to half of \( \sigma_t \).

### 6.3 Examples

Here we list the response times of a few nanoparticles. All of the calculations were
based on solid, metal nanoparticles and their scattering intensity spectra. The re-
sponse time was calculated for spheres, several shapes of the cubic symmetry group,
and crosses. Figure 6.1 shows how the central wavelength and wavelength band-
width were estimated.

**Figure 6.1:** Scattering intensity spectra for a nanoparticle. The response
curve is modelled as Gaussian so that the bandwidth is taken at the FWHM
and the central wavelength corresponds to the position of the peak.

Table 6.1 shows the response times for spheres (see Ref. [48]). The 80 nm silver
nanosphere has the fastest response time, which is attributed to the particle having
the largest bandwidth. When comparing the two silver nanospheres, it is evident
that the response time becomes smaller as the particle size and bandwidth increases. One would initially expect the gold nanosphere to have the fastest response time given that the polarizability increases as you move down a group of the periodic table [53]. However, our formalism does not provide a direct relationship between the polarizability and scattering rate of a particle. The key variables from Eq. (6.6) are the central frequency and frequency bandwidth.

**Table 6.1**

Response time of spheres.

<table>
<thead>
<tr>
<th>Size (nm)</th>
<th>Metal</th>
<th>$\lambda_0$ (nm)</th>
<th>Bandwidth (THz)</th>
<th>Response time (fs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>60</td>
<td>silver</td>
<td>479</td>
<td>460</td>
<td>37</td>
</tr>
<tr>
<td>80</td>
<td>silver</td>
<td>494</td>
<td>720</td>
<td>15</td>
</tr>
<tr>
<td>80</td>
<td>gold</td>
<td>559</td>
<td>490</td>
<td>28</td>
</tr>
</tbody>
</table>

The response times of nanocubes from Ref. [49] and Ref. [50] are displayed in Table 6.2. When comparing the response times of the nanocubes that are approximately 80 nm, we see that the smallest response time is for the silver particle, which is also due to the particle having the largest bandwidth. The 100 nm copper cube has the fastest response time overall; however, the bandwidth of the 100 nm cube is smaller than the bandwidth of the silver nanocube. Based on the trends seen in Table 6.2, it is possible that a 100 nm silver cube will have a larger bandwidth. Thus, the silver nanocube will have a faster response time than the 100 nm copper cube.

**Table 6.2**

Response times of cubes.

<table>
<thead>
<tr>
<th>Size (nm)</th>
<th>Metal</th>
<th>$\lambda_0$ (nm)</th>
<th>Bandwidth (THz)</th>
<th>Response time (fs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>60</td>
<td>copper</td>
<td>615</td>
<td>420</td>
<td>34</td>
</tr>
<tr>
<td>80</td>
<td>copper</td>
<td>641</td>
<td>620</td>
<td>15</td>
</tr>
<tr>
<td>83</td>
<td>silver</td>
<td>450</td>
<td>840</td>
<td>12</td>
</tr>
<tr>
<td>83</td>
<td>gold</td>
<td>565</td>
<td>530</td>
<td>24</td>
</tr>
<tr>
<td>100</td>
<td>copper</td>
<td>673</td>
<td>790</td>
<td>9</td>
</tr>
</tbody>
</table>
We notice that the gold nanoparticles have the smallest bandwidths for both cubic and spherical particles. Furthermore, we see that a silver nanocube has a faster response than a silver nanosphere that is nearly the same size as the cube. Although cubes have a larger dipole moment than spheres [54, 55], we can only link the response time of the silver cube to its large bandwidth.

Table 6.3 lists the response time for a tetrahedron from Ref. [49]. So far, this particle has one of the fastest response times and the largest bandwidth. Yet, the tetrahedron is roughly two times the size of the silver nanocube and nanosphere. If we consider a cube and sphere that is approximately 160 nm, their response times can possibly be much faster than the tetrahedron.

Table 6.3
Respone time of a tetrahedron.

<table>
<thead>
<tr>
<th>Size (nm)</th>
<th>Metal</th>
<th>$\lambda_0$ (nm)</th>
<th>Bandwidth (THz)</th>
<th>Response time (fs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>164</td>
<td>silver</td>
<td>590</td>
<td>870</td>
<td>9</td>
</tr>
</tbody>
</table>

The response time for nanoparticles with geometries similar to a cube are presented in Table 6.4. The scattering properties of the nanoparticles can be found in Ref. [51]. The synthesized particles have multiple geometries, but the main shape is one of the following Archimedean solids: truncated tetrahedron, truncated octahedron, or cuboctahedron. See Fig. 6.2 for a scanning electron microscope (SEM) image of the particles and Fig. 6.3 for images of the Archimedean solids.

Figure 6.2: SEM image of 113 nm silver nanoparticles synthesized using hydrogen reduction method. The nanoparticles are a mixture of truncated tetrahedrons, truncated octahedrons, and cuboctahedrons [56].
Overall, these particles have the fastest response times and the largest bandwidths. The response times increase with the size of the nanoparticle from 75 to 78 nm and 97-105 nm. This is due to an error with estimating the central wavelength and bandwidth (nanometers). Given the variation in sizes in contrast to the other geometries, we assume that the response time may stop decreasing after the particle becomes a certain size. This is due to the small changes in response times for the larger particles.

Table 6.4
Response times of Archimedean solids.

<table>
<thead>
<tr>
<th>Size (nm)</th>
<th>Metal</th>
<th>λ₀ (nm)</th>
<th>Bandwidth (THz)</th>
<th>Response time (fs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>48</td>
<td>silver</td>
<td>440</td>
<td>780</td>
<td>14</td>
</tr>
<tr>
<td>61</td>
<td>silver</td>
<td>450</td>
<td>930</td>
<td>10</td>
</tr>
<tr>
<td>75</td>
<td>silver</td>
<td>470</td>
<td>1200</td>
<td>6</td>
</tr>
<tr>
<td>78</td>
<td>silver</td>
<td>470</td>
<td>1100</td>
<td>7</td>
</tr>
<tr>
<td>92</td>
<td>silver</td>
<td>500</td>
<td>1500</td>
<td>3</td>
</tr>
<tr>
<td>97</td>
<td>silver</td>
<td>500</td>
<td>1600</td>
<td>3</td>
</tr>
<tr>
<td>105</td>
<td>silver</td>
<td>510</td>
<td>1500</td>
<td>3</td>
</tr>
<tr>
<td>113</td>
<td>silver</td>
<td>530</td>
<td>1900</td>
<td>2</td>
</tr>
<tr>
<td>120</td>
<td>silver</td>
<td>550</td>
<td>1900</td>
<td>2</td>
</tr>
<tr>
<td>136</td>
<td>silver</td>
<td>570</td>
<td>2100</td>
<td>2</td>
</tr>
</tbody>
</table>
The polarizability matrix for a cross is isotropic, which makes it a special case since a cross is not apart of the cubic symmetry group. We have included the properties and the response time of a cross in Table 6.5. The cross properties were simulated using Comosol Multiphysics software. The cross has the smallest bandwidth of all of the nanoparticles, but not the slowest response time. This is possibly due to the large central wavelength of the cross. Given the observations above, the response time of a cross can be decreased by fabricating the cross with silver or copper.

<table>
<thead>
<tr>
<th>Size (nm)</th>
<th>Metal</th>
<th>λ₀ (nm)</th>
<th>Bandwidth (THz)</th>
<th>Response time</th>
</tr>
</thead>
<tbody>
<tr>
<td>300×100×30</td>
<td>gold</td>
<td>1500</td>
<td>350</td>
<td>20</td>
</tr>
</tbody>
</table>

We have only put emphasis on the nanoparticles with the fastest response time; however, all the examples of nanoparticles provided above have response times in the femtosecond range. Thus, all of the nanoparticles can be used as scatterers. It is important to note that the response times listed above may not be capable of scattering light for some light sources. This is due to the fact that the response times of the nanoparticles may be too slow. This will lead to improper measurements of the polarization time and polarization length of a beam. Therefore, a beam should be examined with multiple nanoparticles to obtain accurate calculations for the polarization properties of the field. Since each random light source does not have the same polarization dynamics, each source will not necessarily be able to use the same nanoscatterer to characterize its instantaneous polarization state.

In Ref. [35], the polarization times of an amplified spontaneous emission (ASE) source and a dual-laser source are given as 115 fs and 15 fs, respectively. Based on these results, the recommended nanoparticle geometry to use for probes are the Archimedean solids due to their ability to provide the most accurate measurements of the fluctuations in the incident field. For the ASE source any size of the Archimedean solid nanoparticle can be used, but for the dual-laser source it is best to use the Archimedean solids ranging from 92-136 nm. Given the complexity of the Archimedean solids geometry and their fabrication method, an alternative nanoparticle geometry for the ASE source is the cube since these particles are easy.
to fabricate and have fast response times when the size of the nanoparticle is large. There are no additional suggestions for the dual-laser source because none of the other geometries have response times fast enough to follow the instantaneous changes in the polarization of the incident beam.
Experimentally investigating temporal polarization fluctuations has been a challenge due to the limitations of current detectors’ response times. There are some studies that have managed to study polarization dynamics in the time domain (without the problems posed by detectors) by modulating the incident light or using different detection schemes such as two-photon absorption. However, the few experiments that have witnessed polarization dynamics cannot establish where the fluctuations in the electromagnetic field originate. Without knowing where the instantaneous polarization properties begin, it is difficult to state what part of an optical setup is influencing the variations in the electric field, which also hinders the applications of polarization dynamics. Therefore, we have considered a theoretical method that can follow the fluctuations in polarization from a known position of an incident beam.

The measurement technique employs apertureless scanning near field microscopy (SNOM) with nanoparticles that can scatter light with femtosecond scale resolution. To test this approach, we have derived the far field scattered by the nanoparticle and determined the instantaneous polarization properties of the scattered far field. When the polarization correlation function, $\gamma_P(\tau)$, for the far field is constant from $t$ to $t + \tau$, the formula reduces to one. If $\gamma_P(\tau)$ is equal to one, the polarization state of the incident field is constant for a specific time in the far field. Given that the polarization state of the far field is unaltered for periods of time, it is possible for a nanoprobe to follow the instantaneous polarization variations in the electric field for specific points of the incident beam.

To ensure that the nanoprobe has a sufficient response time to scatter light in femtoseconds, we also have developed criteria that is necessary for estimating the re-
response times of isotropic nanoparticles. We determined the polarizability of isotropic nanoparticles by assuming the curves of the nanoparticles’ scattering intensity spectra were Gaussian to approximate the central frequency and frequency bandwidth, which assisted in calculating the electric susceptibility. The response times of the nanoparticles are half of the bandwidth of the electric susceptibility. The results show that all nanoparticles considered have response times within the femtosecond range. We see that more complex cubic nanostructures (Archimedean truncated octahedron, Archimedean truncated tetrahedron, and Archimedean cuboctahedron) have the fastest response times, which range from 2-14 fs, and will provide the most accurate representation of the fluctuations in the incident electric field.

A shortcoming for our method is that the setup will not provide any information on the instantaneous properties of left- and right-circularly polarized light. Despite this small drawback, we still believe that this measurement technique can be used for further experiments on temporal polarization dynamics. The proposed approach can be combined with previous techniques for measuring instantaneous polarization properties to review the results for the polarization time. Also, this theory can be expanded to study the polarization fluctuations of near fields.


The electric field scattered from the electric dipole is given by

\[ E = \nabla (\nabla \cdot \Pi_e) - \frac{1}{c^2} \ddot{\Pi}_e. \quad (A.1) \]

To simplify this equation, we first calculate the divergence of the electric Hertz vector:

\[ \nabla \cdot \Pi_e = \nabla \cdot \left( \frac{[p(t)]}{R} \right) \quad (A.2) \]

\[ = \frac{\partial}{\partial x} \left( \frac{p_x(t)}{R} \right) + \frac{\partial}{\partial y} \left( \frac{p_y(t)}{R} \right) + \frac{\partial}{\partial z} \left( \frac{p_z(t)}{R} \right) \]

\[ = - \left( \frac{x[p_x(t)]}{R^3} + \frac{x[\dot{p}_x(t) \cdot c]}{cR^2} \right) - \left( \frac{y[p_y(t)]}{R^3} + \frac{y[\dot{p}_y(t) \cdot c]}{cR^2} \right) - \left( \frac{z[p_z(t)]}{R^3} + \frac{z[\dot{p}_z(t) \cdot c]}{cR^2} \right) \]

\[ = - \left\{ \frac{[p(t)]}{R^3} + \frac{[\dot{p}(t) \cdot c]}{cR^2} \right\} \cdot R. \]
Next, we calculate the gradient of Eq. (A.2) focusing only on \( x \). The calculations are the same for \( y \) and \( z \) due to symmetry.

\[
\nabla(\nabla \cdot \Pi_e) = \nabla \left( - \left\{ \frac{[p(t)]}{R^3} + \frac{[\dot{p}(t)]}{cR^2} \right\} \cdot \mathbf{R} \right) = -\frac{\partial}{\partial x} \left[ \left( \frac{[p_x(t)]}{R^3} + \frac{[\dot{p}_x(t)]}{cR^2} \right) x + \left( \frac{[p_y(t)]}{R^3} + \frac{[\dot{p}_y(t)]}{cR^2} \right) y \right. \\
\left. + \left( \frac{[p_z(t)]}{R^3} + \frac{[\dot{p}_z(t)]}{cR^2} \right) z \right] + \ldots \\
= \left( \frac{3x[p_x(t)] + 3y[p_y(t)] + 3z[p_z(t)]}{R^3} + \frac{3x[\dot{p}_x(t)] + 3y[\dot{p}_y(t)] + 3z[\dot{p}_z(t)]}{cR^2} \\
+ \frac{x[\ddot{p}_x(t)] + y[\ddot{p}_y(t)] + z[\ddot{p}_z(t)]}{c^2R^3} \right) R - \left\{ \frac{[p(t)]}{R^3} + \frac{[\dot{p}(t)]}{cR^2} \right\} \cdot \mathbf{R}.
\]

(A.3)

Now we calculate the second derivative of the Hertz derivate with respect to time:

\[
\dddot{\Pi}_e = \frac{\partial^2}{\partial t^2} \left[ \frac{[p(t)]}{R} \right] = \frac{[\dddot{p}(t)]}{R}.
\]

(A.4)

Substituting Eq. (A.3) and Eq. (A.4) into Eq. (A.1), the scattered electric field becomes

\[
E = \left( \left\{ \frac{3[p(t)]}{R^5} + \frac{3[\dot{p}(t)]}{cR^4} + \frac{[\dddot{p}(t)]}{c^2R^3} \right\} \cdot \mathbf{R} \right) R - \left\{ \frac{[p(t)]}{R^3} + \frac{[\dot{p}(t)]}{cR^2} + \frac{[\dddot{p}(t)]}{c^2R} \right\} \cdot \mathbf{R}.
\]

(A.5)

The scattered electric field presented in Eq. (A.5) is for any observation direction. We can further simplify the equation for the scattered electric field for our observation direction by expanding the polarization vector:

\[
[p(t)] = \frac{p_x(t)(\hat{u}_R + \hat{u}_\theta)}{\sqrt{2}} + \frac{\sqrt{2}p_y(t)\hat{u}_z}{\sqrt{2}} + \frac{p_z(t)(\hat{u}_R - \hat{u}_\theta)}{\sqrt{2}}.
\]

(A.6)
Given that the electric field is only polarized in the $x$- and $y$-direction, Eq. (A.6) can be written as

$$[p(t)] = \frac{[p_x(t)(\hat{u}_R + \hat{u}_\theta)]}{\sqrt{2}} + \frac{\sqrt{2}p_y(t)\hat{u}_\varphi}{\sqrt{2}} \tag{A.7}$$

$$= \frac{[p_x(t)(\hat{u}_R + \hat{u}_\theta) + \sqrt{2}p_y(t)\hat{u}_\varphi]}{\sqrt{2}}. \tag{A.7}$$

Using Eq. (A.7) and recalling the definitions of $R$ and the spherical polar coordinate system unit vectors [see Eq. (5.30) and Eq. (5.34)--(5.36)], the electric scattered field is

$$E = \left\{ \frac{3[p_x(t)]}{\sqrt{2}R^4} + \frac{3[\dot{p}_x(t)]}{\sqrt{2}cR^3} + \frac{[\ddot{p}_x(t)]}{\sqrt{2}c^2 R^2} \right\} R - \left\{ \frac{[p(t)]}{R^3} + \frac{[\dot{p}(t)]}{cR^2} + \frac{[\ddot{p}(t)]}{c^2 R} \right\}. \tag{A.8}$$