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Research Paper

Astrobiological Polarimeter

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Abstract

Chirality is an excellent indicator of life, but naturally occurring astrobiological (as well as terrestrial) samples nearly always exhibit massive depolarizing light scattering, which renders conventional polarimeters useless. For astrobiological applications, we instead consider a novel polarimeter originally developed for non-invasive human-glucose measurement. It involves deliberately rotating in time the plane of polarization of a linearly polarized beam and detecting the shift in the plane of the rotating linearly polarized component of the transmitted light from a chiral sample relative to the input polarization plane. We find that this polarimeter can operate in 3 orders of magnitude more depolarizing scattering than conventional polarimeters. Furthermore, it can also be designed to be lightweight, compact, and energy efficient. Key Words: Chirality—Polarimeter—Scattering—Depolarization. Astrobiology 8, 1061–1069.

Introduction: Life, Chirality, and Its Measurement

Since Pasteur's seminal observations with enantiomers of tartrate over one hundred fifty years ago, chirality has been recognized as playing a critical role in biological systems. Whereas the chemistry of inanimate systems rarely shows chiral preference, chirality is commonplace and often quite strong in biological systems (Bonner, 1995). Nearly all biological polymers must be homochiral (all its component monomers having the same handedness, *i.e.*, the same enantiomer) to function (MacDermott *et al.*, 1996; Wang, 1997). For example, all amino acids in proteins are "left-handed," while all carbohydrates (sugars) in DNA, RNA, and the metabolic pathways are "right-handed." Thus, the detection of chirality is an excellent indicator of extraterrestrial life (MacDermott *et al.*, 1996; Thaler *et al.*, 2006).

Current chiral detectors, which include high-pressure liquid chromatographs, gas chromatographs, and capillary electrophoresis, are definitive in identifying specific chiral species, for which they use a compound-specific standard. For space missions looking for signs of life—that is, chirality in general—these aforementioned techniques would likely miss unanticipated chiral species of exotic life-forms. A polarimeter, on the other hand, detects chirality in general by measuring the polarization rotation produced by any chiral compound. So while a polarimeter does not identify the specific chiral compound, it is still best suited for finding the initial signs of life in dissolved samples. It should be mentioned, of course, that some birefringent minerals (crystals) are liquid, *e.g.*, water soluble, and in solution exhibit optical rotation due to their birefringence. This could be confused by a polarimeter. In any case, devices with increased specificity would then be appropriate in subsequent missions to conduct more qualitative investigations at the same location.

For terrestrial use, polarimeters are quite common, all of which essentially comprise pairs of crossed polarizers with assorted modulators and detectors (Westbrook *et al.*, 2000; Hirabayashi and Amano, 2003; Temporao and Von der Weid, 2003; Chou *et al.*, 2006). However, little effort has been devoted to polarimeters for astrobiological purposes during space exploration. We know of only one such effort, the "SETH-Cigar," which was developed in Europe for Mars and

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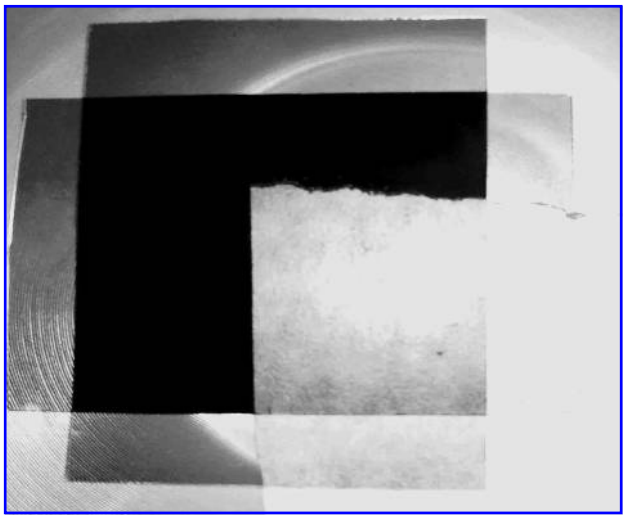

FIG. 1. A piece of wax paper (lower right) held between crossed polarizers depolarizes the light and causes significant light to pass through the second polarizer.

other extraterrestrial destinations; it comprises a carefully engineered compact pair of crossed polarizers (MacDermott *et al.*, 1996).

Unfortunately, naturally occurring samples on Earth and elsewhere, whether living or nonliving, exhibit significant depolarizing light scattering (DLS). DLS occurs because naturally occurring matter is structurally complex and has many tiny regions of different refractive indices and absorption coefficients. In addition to scattering the beam, each of these individual regions absorbs, reflects, refracts, or phase-delays orthogonal polarizations by different amounts. DLS does not require that the medium or its small subregions have any birefringence or dichroism (the tendency for one polarization to be absorbed more than its orthogonal counterpart), both of which are well known to rotate polarization, though, of course, these properties induce additional depolarization. A light ray propagating through a large number of such tiny regions evolves to an arbitrary polarization state. Thus a light beam, which consists of multiple rays of light that experience significant DLS, develops a random polarization distribution in space; and its transmission cannot be blocked by any orientation of a polarizer. This is illustrated by a simple Polaroid–wax paper experiment (Fig. 1). Unpolarized light from a lamp becomes linearly polarized upon transmission through the Polaroid sheet, and a second Polaroid in the crossed orientation blocks the transmission of the incident orthogonally polarized light. However, wax paper inserted between the crossed polarizers introduces DLS and, hence, depolarizes the light (creates a random spatial polarization distribution). This results in a dramatic increase in transmission through the second Polaroid-sheet polarizer.

The degradation of the polarization of a light beam experiencing DLS is further illustrated in Fig. 2, which shows a significant decrease in the *degree of polarization* of transmitted light (the ratio of transmitted light power measured at transmission-maximum and transmission-minimum of a polarizer) with increased DLS. As a result, the performance of conventional polarimeters deteriorates badly in the presence of DLS (Nee and Cole, 1998; Chaikovskaya and Zege, 1999). Consequently, conventional polarimeters require the user to prepare "optically clean" samples, that is, samples essentially free of scattering structures and its resulting DLS. This means the removal of essentially all particulate matter from the sample surface and interior, which leaves a clear, homogenous liquid sample—something utterly impractical in extraterrestrial environments. Thus, conventional polarimeters are ineffective for astrobiological applications. Only polarimeters that measure the complete polarization state of light, called Stokes vector polarimeters, are able to handle depolarized light. However, the DLS in a practical sample

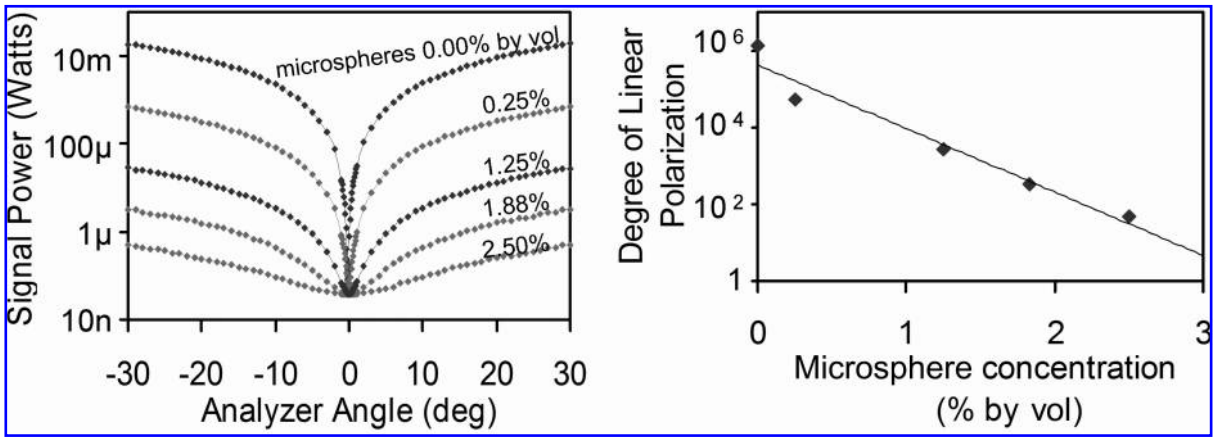

FIG. 2. **(**Left) Malus Law behavior of the light power transmitted through a polarizer pair with samples containing several different concentrations of 10 μm polystyrene microspheres added to water placed in between. In the absence of DLS (0% spheres), there is a sharp decrease in transmitted power when the polarizers are crossed (analyzer angle = 0°). However, increased concentration of microspheres reduces the difference between the light power measured at transmission maximum and minimum. (**Right**) The degree of linear polarization of transmitted light from samples with different microsphere concentrations (the ratio of the light power measured at transmission maximum when the polarizers are parallel and transmission minimum when the polarizers are crossed), or equivalently, the polarizer extinction ratio.

can induce a large depolarized component that will easily overwhelm the polarized component, and a more sensitive polarimeter is required to extract the chirality information buried under the massive depolarized background.

To illustrate this problem—in a very terrestrial setting—we consider an attempt to determine whether a *human* is a living organism solely by detecting the subject's chirality by way of a conventional polarimeter. This would seem a simple problem in view of a typical human's significant amount, ~1g/L, of glucose, a molecule with one of the highest known specific optical rotations ($[\alpha]_D^{24} = 52.5°$). Unfortunately, the extremely high DLS in human tissue would overwhelmingly depolarize the light beam, and the polarized signal component that carries information about the little amount of sugar present in the tissue would be lost to the background. Thus, a conventional polarimeter fails badly for such a sample.

Indeed, the problem of detecting astrobiological chirality bears a striking resemblance to that of developing a chirality-based non-invasive human glucose monitor for diabetics, who must frequently monitor their glucose levels. Both applications require a lightweight, compact, power-efficient, and robust polarimeter; most importantly, both require the measurement of chirality in the presence of significant amounts of DLS (Atkins and Barron, 1970; Mackintosh *et al.*, 1989; Toropainen, 1993; Marienko and Savenkov, 1994; Barry *et al.*, 1997; Delplancke *et al.*, 1997; Vitkin and Hoskinson, 2000; Chaikovskaya, 2002). In fact, the non-invasive human-glucose monitoring problem is actually more difficult; it also requires low-cost manufacturability, specificity for glucose, and approximately 90% accuracy. No such stringent conditions are required for a chirality monitor for astrobiological applications, where only a few devices are likely to be constructed, *any* chiral substance detected would be interesting, and an order-of-magnitude result would suffice.

Unfortunately, a chirality-based non-invasive human-glucose monitor remains an unsolved problem (Cote, 1997). Much effort has been expended on it, however, and astrobiology would benefit from the experience of this disparate community (Cameron and Cote, 1997; Cote, 1997; Cote and Cameron, 1997; Cameron *et al.*, 2000; McShane *et al.*, 2000; Baba and Cote, 2002). So, for astrobiological applications, we have investigated a chirality monitor that was first developed for non-invasive *in vivo* human glucose sensing (Kupershmidt, 1995a, 1995b, 1997; Kupershmidt *et al.*, 1996) but abandoned due to insufficient accuracy (R. Tillman, 2006, private communication).

This polarimeter involves continuously rotating the plane of linear polarization of a laser beam, which then passes through a sample with DLS. The transmitted beam, when analyzed with a fixed-orientation analyzer, generates a sinusoidal voltage signal. It then compares this signal with a voltage signal for an analogous setup without a sample. If the sample medium is chiral, it further rotates the (linear) polarization of the beam, which introduces a *phase difference* between its sinusoidal output voltage and that of the reference beam. This phase difference indicates the sample chirality.

Method: The Rotating Polarization Polarimeter

Polarimeters determine the chirality of a sample medium by measuring the amount of polarization rotation induced by a sample on the light propagating through it. If one linear polarizer generates a purely linear polarization, the transmittance T of a second linear polarizer (called an analyzer) is given by Malus law, $T = \cos^2(\delta)$, where δ is angle between the plane of polarization of incident linearly polarized light and the transmission axis of the analyzer. The sample medium causes rotation of the polarization by an angle (α) proportional to the sample's chirality. The determination of chirality requires finding the *phase shift* generated in the transmitted intensity sinusoid upon introduction of a chiral sample, as shown in Fig. 3. This can be done by rotating the analyzer to find the new angle of minimum transmission.

Conventional polarimeters simply use a good set of polarizers (extinction ratio ~10^6:1) and a photo detector that allows for a quantitative measurement of intensity to determine α (Oliva, 1997). Recently proposed polarimetry techniques utilize polarization- or intensity-modulated input light, optical heterodyne detection, advanced electronics, and post-measurement data processing with lock-in detection techniques to determine the polarization rotation with higher sensitivity (Goldstein, 1992; Chou *et al.*, 1997; Berezhna *et al.*, 2001; Blakeney *et al.*, 2002).

Polarimeters based on modulating the input polarization (*i.e.*, the optical phase) generally dither the instantaneous linear polarization by a few degrees about a mean linear polarization. The signal amplitudes measured at various harmonics of the modulation frequency are then used to determine the sample-induced polarization rotation. Some techniques require measurements for different mean linear polarizations. The performance (sensitivity and accuracy) of all of these techniques, however, deteriorates badly in the presence of DLS, which depolarizes the light and, hence, severely reduces the amplitude of the output signal-voltage sinusoid.

Cote *et al.* (1992) proposed a true phase-shift measurement technique that overcomes the amplitude noise effects (Cote

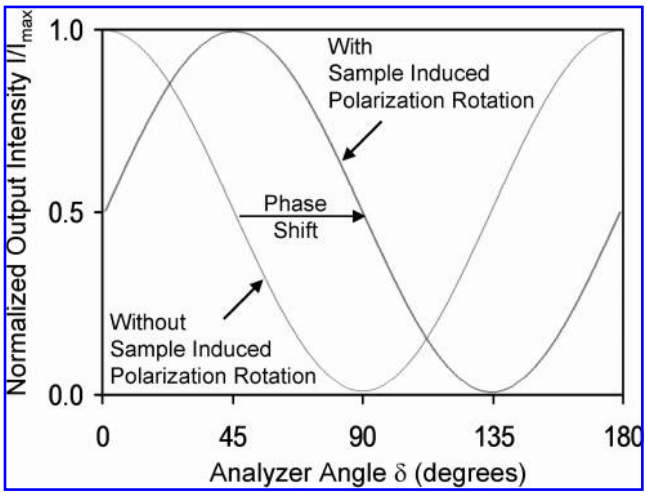

FIG. 3. Transmittance of a linear polarizer (from Malus law) expressed as normalized output intensity (I/I_{max}) vs. δ, the angle between the incident plane of polarization and the transmission axis of the polarizer. Propagation through a chiral sample rotates the linear polarization, which manifests itself as a phase-shifted sinusoidal transmission curve.

et al., 1992). Here, we further investigate this technique for its potential to overcome significant DLS. The technique involves lock-in detection of signal *phase* (not amplitude) of the output voltage acquired from a rotating linear polarization, which we briefly described in the previous section and will describe in more detail in the next section. We refer to this as the rotating-polarization (RP) polarimeter. Cote *et al.* (1992) achieved rotating linear polarization with the use of a linear polarizer and a quarter-wave plate (QWP) to first yield a circularly polarized beam and a rotating linear polarizer. While this sequence of optics allows the use of multiple wavelengths (polarizers are broadband), it results in the loss of half the intensity of light provided by the first polarizer. To achieve rotating linear polarization in our RP polarimeter, we use instead a linear polarizer and a rotating QWP/mirror combination [this combination effectively behaves as a half-wave plate (HWP)], which minimizes the intensity loss in the process and yields a more energy-efficient device.

While DLS decreases both the dc- and ac-components of the transmitted light and voltage signals, *it does not affect the detected voltage phase difference induced by chirality*. A lock-in amplifier then extracts the relative phase between the signal and reference voltage sine waves. Lock-in detectors are known for their incredible sensitivity; they can measure a sine wave component buried in over 100 dB of noise. This is ideal for tiny chirality signals buried in massive DLS. The experimental system utilizes lock-in detection, and the complete setup is illustrated in Fig. 4.

Theory

For simplicity, we first analyze the polarimeter response to an *optically clean sample*. Since Jones calculus can be used to analyze fully polarized light, we use it to derive expressions for the output light signals of the RP polarimeter (Rochford, 2004). We assume that the samples do not exhibit any circular dichroism or absorption at the experimental wavelength, so we ignore these effects. We also assume that the sample does not exhibit any linear birefringence.

A collimated laser beam is split into two, and both beams are passed through a polarizer to create two vertically polarized beams, which act as the reference and sample signal beams. The *E*-fields for both beams have the following Jones vectors:

$$\mathrm{E}_{\text{Reference Beam}} = \mathrm{E}_{\text{in}}\begin{bmatrix}0\\1\end{bmatrix};\ \mathrm{E}_{\text{Sample Beam}} = \mathrm{E}_{\text{in}}\begin{bmatrix}0\\1\end{bmatrix} \qquad [1]$$

A zero-order quarter-wave plate (QWP) and a mirror then act as a half-wave plate (HWP), which rotates the polarization of both the beams by an identical amount: 2θ, where θ is the rotation angle of the QWP. Mechanically rotating the QWP/mirror at a frequency ω then rotates the polarization of the 2 beams at twice the mechanical frequency, 2ω. Since $\theta = \omega t$, where t is time, the Jones matrix for combination of QWP oriented at an angle θ from vertical and the mirror, and the resultant Jones vectors for the reference and signal beam with rotating polarizations are

$$\text{Rotating}_{\text{QWP,Mirror}} = e^{i\frac{\pi}{2}}\begin{bmatrix}\cos(2\omega t) & -\sin(2\omega t)\\ \sin(2\omega t) & \cos(2\omega t)\end{bmatrix} \qquad [2]$$

$$\mathrm{E}^{\text{Rotating}}_{\text{Reference Beam}} = \text{Rotating}_{\text{QWP,Mirror}} \cdot \mathrm{E}_{\text{in}}\begin{bmatrix}0\\1\end{bmatrix} = \mathrm{E}_{\text{in}}\, e^{i\frac{\pi}{2}}\begin{bmatrix}-\sin(2\omega t)\\ \cos(2\omega t)\end{bmatrix} \qquad [3]$$

$$\mathrm{E}^{\text{Rotating}}_{\text{Sample Beam}} = \mathrm{E}_{\text{in}}\, e^{i\frac{\pi}{2}}\begin{bmatrix}-\sin(2\omega t)\\ \cos(2\omega t)\end{bmatrix} \qquad [4]$$

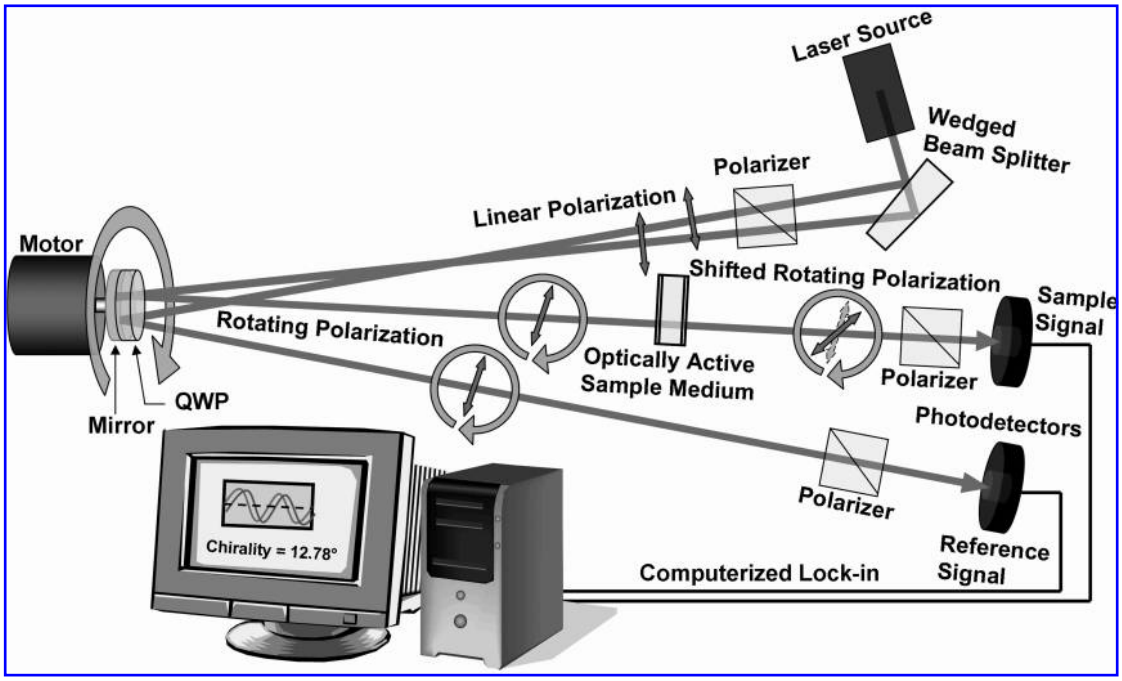

FIG. 4. Rotating polarization (RP) polarimeter setup.

The reference beam then propagates through a vertical polarizer to a photodetector that produces a sinusoidal voltage signal, $V_{\text{Reference}}$.

$$\mathrm{E}_{\text{Output Reference Beam}} = \begin{bmatrix} 0 & 0 \\ 0 & 1 \end{bmatrix} \cdot \mathrm{E}^{\text{Rotating}}_{\text{Sample Beam}} = \mathrm{E}_{\text{in}} e^{i\frac{\pi}{2}} \begin{bmatrix} 0 \\ \cos(2\omega t) \end{bmatrix} \quad [5]$$

$$\therefore \text{Intensity}_{\text{Output Reference Beam}} \propto |\mathrm{E}_{\text{Output Reference Beam}}|^2$$

$$\because \mathrm{V}_{\text{Reference}} \propto \text{Intensity}_{\text{Output Reference Beam}} \propto \cos^2(2\omega t) = \frac{1}{2}\cos(4\omega t) + \frac{1}{2} \quad [6]$$

The sample signal beam propagates through the sample medium, which rotates the plane of polarization of the beam by an angle α proportional to the chirality of the sample medium. Consequently, the plane of polarization of the signal beam still rotates at the same frequency 2ω; but, due to sample chirality, it now develops a *phase difference* α with respect to the reference beam. The signal beam then propagates through a polarizer to the photodetector, which then produces a phase-shifted voltage signal, V_{Sample}.

$$\mathrm{E}_{\text{Output Reference Beam}} = \begin{bmatrix} 0 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} \cos(\alpha) & -\sin(\alpha) \\ \sin(\alpha) & \cos(\alpha) \end{bmatrix} \mathrm{E}^{\text{Rotating}}_{\text{Sample Beam}} \quad [7]$$

$$\therefore \mathrm{E}_{\text{Output Sample Beam}} = \mathrm{E}_{\text{in}} e^{i\frac{\pi}{2}} \begin{bmatrix} 0 \\ \cos(2\omega t + \alpha) \end{bmatrix} \quad [8]$$

$$\because \text{Intensity}_{\text{Output Sample Beam}} \propto |\mathrm{E}_{\text{Output Sample Beam}}|^2$$

$$\therefore \mathrm{V}_{\text{Sample}} \propto \text{Intensity}_{\text{Output Sample Beam}} \propto \cos^2(2\omega t + \alpha) = \frac{1}{2}\cos(4\omega t + 2\alpha) + \frac{1}{2} \quad [9]$$

The reference and sample voltage signals are then processed by a computerized lock-in detector, which measures the phase difference Φ between the sample and reference voltage signal waves at frequency 4ω, which represents the polarization rotation ($\Phi = 2\alpha$ as evident from expressions for $V_{\text{Reference}}$ and V_{Sample}).

The preceding analysis assumes an optically clean sample, which introduces only optical rotation and does not exhibit any DLS. Now we examine the effect of non-optically clean samples, which also exhibit DLS. Multiple scattering is an extensively studied phenomenon in, for example, the bioimaging community (Cote and Vitkin, 2004; Swami *et al.*, 2006). Multiple scattering in the Rayleigh regime (when the size of scattering particles is much smaller, ~1/10 or less, than the wavelength of scattered radiation) and the Mie regime (scattering by larger particles) is characterized through the complex Mueller matrix for emerging radiation in the forward-scattering as well as back-scattering direction (Mueller and Crosbie, 2000; Nee, 2001; Grin'ko and Shkuratov, 2002; Manhas *et al.*, 2006). The transmitted beam from a sample with DLS has both ballistic (*i.e.*, unscattered, hence polarized) and incoherent (*i.e.*, scattered, hence depolarized) components. In the rotating-polarization polarimeter, the incoherent (depolarized) component of light contributes a dc signal while reducing as well the signal strength (intensity, and therefore the voltage amplitude) of the ac polarized component, which still rotates at the same frequency, with its phase shifted by an amount equal to the chirality of the sample.

The above effects are described by Mueller matrices as follows: The Stokes vector describing both the vertically polarized input sample and reference beams are

$$\mathrm{S}_{\text{Reference Beam}} = \mathrm{S}_0 \begin{bmatrix} 1 \\ 1 \\ 0 \\ 0 \end{bmatrix}; \mathrm{S}_{\text{Sample Beam}} = \mathrm{S}_0 \begin{bmatrix} 1 \\ 1 \\ 0 \\ 0 \end{bmatrix} \quad [10]$$

The reference and sample beams propagate through the QWP/mirror combination (effectively a HWP), rotating at a frequency ω. The Mueller matrix that describes the QWP/mirror combination rotating at a frequency ω and the resultant Stokes vectors for the reference and signal beam with rotating polarization are:

$$\text{Rotating}_{\text{QWP/Mirror}} = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos(4\omega t) & \sin(4\omega t) & 0 \\ 0 & -\sin(4\omega t) & \cos(4\omega t) & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix} \quad [11]$$

$$\mathrm{S}^{\text{Rotating}}_{\text{Reference Beam}} = \text{Rotating}_{\text{QWP/Mirror}} \cdot \mathrm{S}_0 \begin{bmatrix} 1 \\ 1 \\ 0 \\ 0 \end{bmatrix} = \mathrm{S}_0 \begin{bmatrix} 1 \\ \cos(4\omega t) \\ -\sin(4\omega t) \\ 0 \end{bmatrix} \quad [12]$$

$$\mathrm{S}^{\text{Rotating}}_{\text{Sample Beam}} = \mathrm{S}_0 \begin{bmatrix} 1 \\ \cos(4\omega t) \\ -\sin(4\omega t) \\ 0 \end{bmatrix} \quad [13]$$

The reference beam propagates through a vertical analyzer to the photodetector, which results in an output Stokes vector, $S_{\text{Output Reference Beam}}$.

$$\mathrm{S}_{\text{Output Reference Beam}} = \frac{1}{2} \begin{bmatrix} 1 & -1 & 0 & 0 \\ -1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix} \cdot \mathrm{S}^{\text{Rotating}}_{\text{Reference Beam}} \quad [14]$$

$$\therefore \mathrm{S}_{\text{Output Reference Beam}} = \frac{1}{2}\mathrm{S}_0\{1 - \cos(4\omega t)\} \begin{bmatrix} 1 \\ -1 \\ 0 \\ 0 \end{bmatrix} \quad [15]$$

The Mueller matrix (M_{Sample}) that describes a chiral sample medium, which also exhibits DLS, can be expressed as the sum of a non-scattering polarization-rotating matrix and a scattering depolarizing matrix. These individual matrices describe the effect of a scattering chiral sample medium on the input radiation. Specifically, the ballistic (unscattered) light only undergoes polarization rotation and constitutes the nondepolarized rotation matrix. On the other hand, multiply scattered light (in the forward direction) becomes completely depolarized and incoherent. We represent the ballistic light contribution to the normalized total signal by β, so the contribution of depolarized component is $(1 - \beta)$:

$$M_{Sample} = \beta \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos(2\alpha) & -\sin(2\alpha) & 0 \\ 0 & \sin(2\alpha) & \cos(2\alpha) & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix} + (1-\beta)\begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix} \quad [16]$$

$$\therefore M_{Sample} = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & \beta\cos(2\alpha) & -\beta\sin(2\alpha) & 0 \\ 0 & \beta\sin(2\alpha) & \beta\cos(2\alpha) & 0 \\ 0 & 0 & 0 & \beta \end{bmatrix} \quad [17]$$

The sample beam propagates through the sample medium that also exhibits DLS and then a vertical analyzer to the photodetector, which results in a voltage signal that is proportional to $S_{Output\ Sample\ Beam}$.

$$S_{Output\ Sample\ Beam} = \frac{1}{2}\begin{bmatrix} 1 & -1 & 0 & 0 \\ -1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix} \cdot M_{Sample} \cdot S^{Rotating}_{Sample\ Beam} \quad [18]$$

$$\therefore S_{Output\ Sample\ Beam} = \frac{1}{2} S_0\{1 - \beta\cos(4\omega t + 2\alpha)\}\begin{bmatrix} 1 \\ -1 \\ 0 \\ 0 \end{bmatrix} \quad [19]$$

Analyzing the Stokes vectors for the output beams confirms that the phase difference between the 2 signals is the same 2α as in the case of no DLS at frequency 4ω. Therefore, the RP polarimeter appears to be an excellent method for measuring rotation induced by a chiral sample, undeterred by DLS.

Results

To demonstrate that the polarimeter works in the absence of DLS, we used the RP polarimeter to measure several optically clean samples (*i.e.*, without DLS) with varying concentrations of glucose and fructose. The measured polarization rotation changed linearly upon varying the concentration of chiral solutes (glucose and fructose), which verified the integrity of the device (Fig. 5).

We used 2% homogenized cow's milk as a source of DLS. Milk is a natural substance that exhibits a considerable amount of scattering, which is responsible for milk's opaque white appearance. Milk also mixes homogeneously with water solutions of fructose and glucose. Figure 6 shows a comparison between the response of the RP polarimeter (dashed curves) with that of a conventional polarimeter (solid curves) in measuring the polarization rotation caused by water and solutions of glucose (1 *M*) and fructose (1 *M*) in the presence of increasing amounts of milk. Note that the conventional polarimeter fails to determine the polarization rotation in the presence of more than 5% milk (by volume), whereas the RP polarimeter accurately detects the polarization rotation in the presence of up to 20% milk.

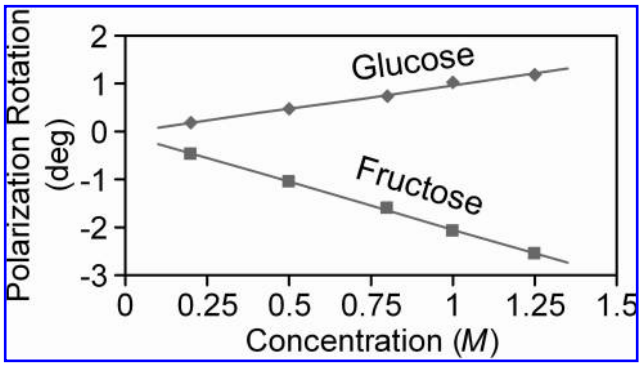

FIG. 5. The RP polarimeter produces the appropriate linear response of polarization rotation to the increasing concentrations of glucose and fructose.

Discussion

Figure 7 provides more detail on the RP polarimeter's immunity to significant DLS, associating the DLS magnitude for a given concentration of milk. While the lock-in phase determines the polarization rotation, the lock-in amplitude is a measure of the fraction of unscattered light (Eq. 19). So, in Fig. 7, we illustrate the normalized lock-in amplitude vs. milk concentration (solid line). A fourfold increase in the concentration of milk (from 5% to 20%) corresponds to three orders of magnitude decrease in the fraction of unscattered light, an additional measure of this polarimeter's capabilities. Thus, the RP polarimeter significantly outperforms conventional polarimeters by three orders of magnitude of DLS.

In addition, Fig. 7 shows the calculated fraction of unscattered light estimated by Beer-Lambert's Law (dashed line), *i.e.*, an exponential decrease of the intensity of unscattered light with distance and scatterer concentration. Note that the actual fraction of photons that remained unscattered at larger milk concentrations were measurable and, thus, greater than the estimation from Beer-Lambert's Law due to a variety of complex effects [it is known that Beer-Lambert's Law breaks down for large concentrations of scatterers (Schnorrenberg *et al.*, 1995)].

The use of a lock-in detector to extract the phase of a signal component at any reference frequency (here 4ω) provides a significant dynamic range for measurement (over 8 orders of magnitude change in the signal amplitude in our setup). The sensitivity and accuracy of the detector in measuring the phase remains roughly constant throughout this range, and these numbers deteriorate only when the amplitude of the desired signal component at the reference frequency is reduced to less than 10^{-6} of the total signal. The sources of noise are the unwanted forward scattered signal, rotational frequency instability of the motor, and the inherent noise in the electrical components of the setup.

Our RP polarimeter can measure a variation of less than 0.1° of polarization rotation in the presence of more than 15% milk (by volume), with standard deviations in the measurement of the phase values of up to ±0.035° (Fig. 8). For *in vitro* glucose-sensing applications, the sensitivity of our device is ~20g/L ± 7g/L for a 1 cm long path length sample cell. Thus, this polarimeter is not sensitive enough to measure

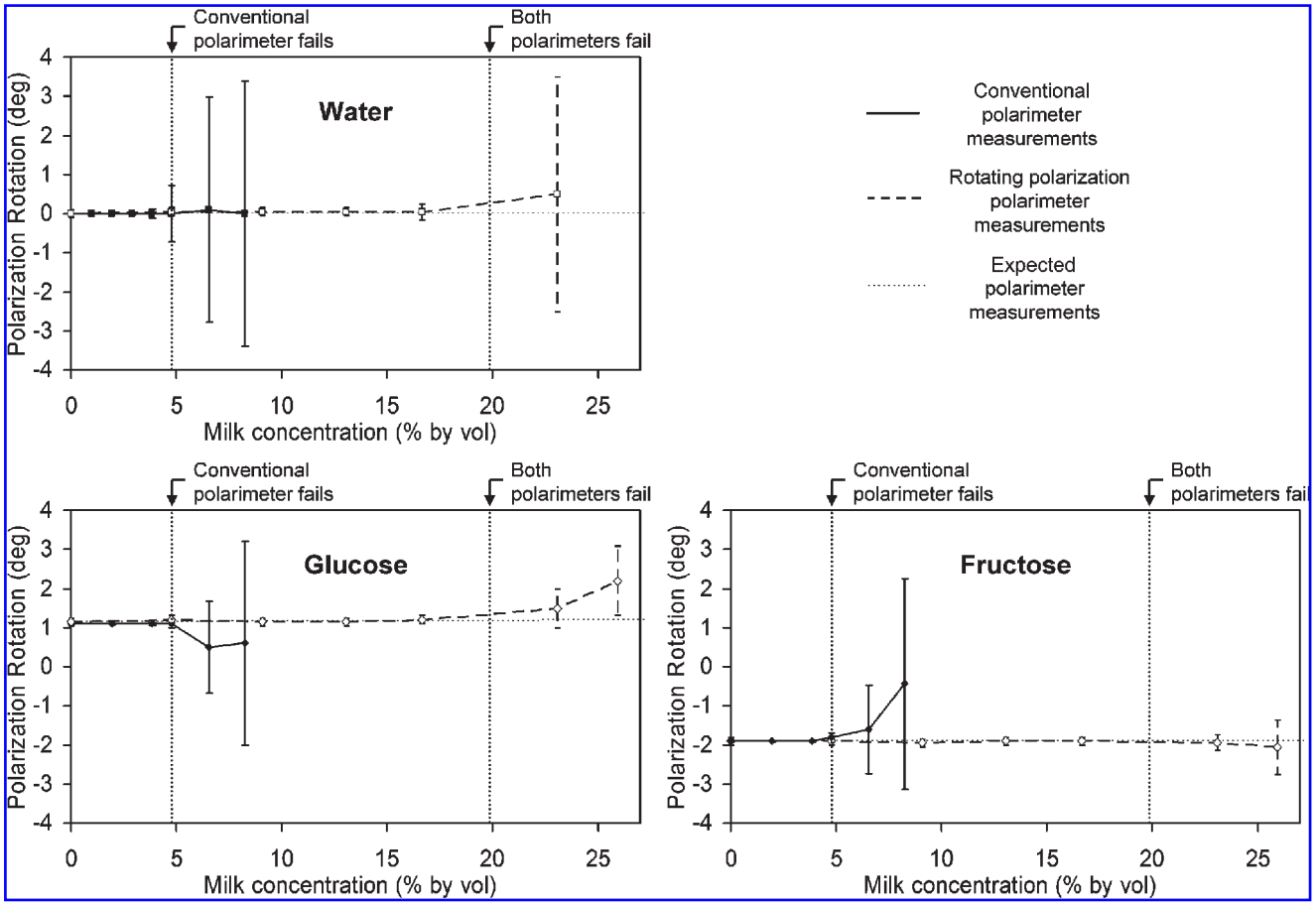

FIG. 6. Measurements of polarization rotation by water, fructose, and glucose solutions, with a conventional polarimeter and a RP polarimeter, in the presence of milk (2% homogenized cow's milk, Kroger Dairy). The increasing size of error bars show that the conventional polarimeter fails to accurately measure the polarization rotation in the presence of 5% milk or more, whereas the RP polarimeter continues to accurately measure polarization rotation for up to the presence of 20% milk.

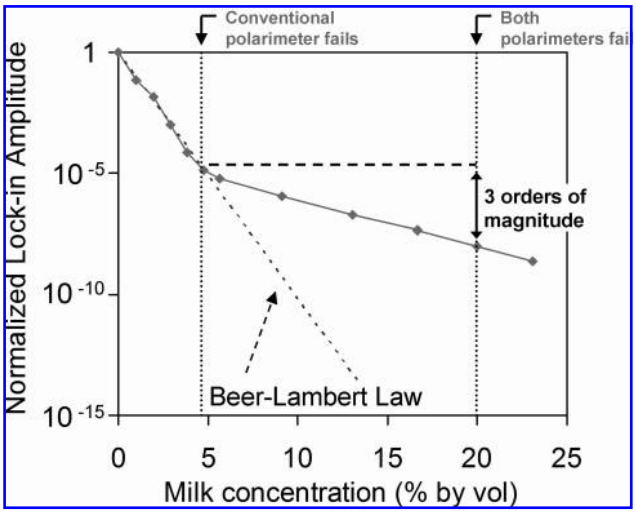

FIG. 7. Solid curve: Normalized lock-in amplitude as measured by the RP polarimeter vs. milk concentration. Dashed curve: Estimated fraction of unscattered light from Beer-Lambert's Law.

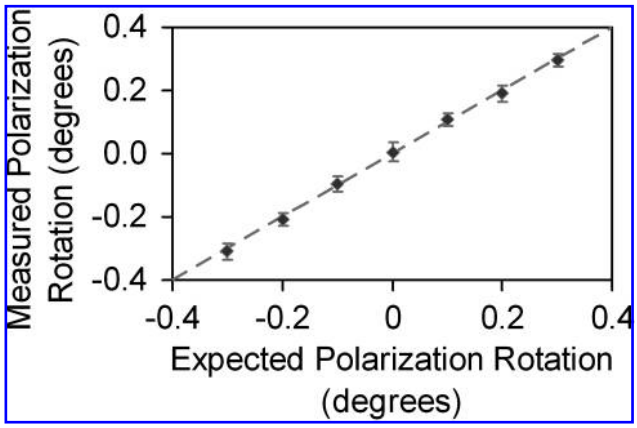

FIG. 8. The RP polarimeter accurately measures a variation in polarization rotation smaller than 0.1° in the presence of 15% milk (2% homogenized cow's milk, Kroger Dairy), with standard deviations up to ±0.035°. The accuracy of the RP polarimeter was tested with samples of low-concentration fructose and glucose solutions mixed with milk, which thus exhibit massive DLS.

typical blood glucose values in humans. Its sensitivity could be increased further by improving the mechanical stability of the motor and the QWP/mirror attached to it, so that a more stable mechanical rotation frequency could be achieved.

We note that some constituents of milk are also chiral in nature, mostly lactose, which constitutes ~5% of milk (~12 g per 240 ml) (Chandan, 1997). Since the specific rotation of lactose is 55°/(g/ml)/dm, we expect 100% milk to rotate the polarization of linearly polarized light in a 1 cm long sample cell by 0.275°. The maximum concentration of milk in our solutions was 25%, which means that the contribution of milk to the total polarization rotation was always below 0.07°. We were unable to see the small contribution of milk to overall polarization rotation in our experiments due to insufficient sensitivity in our current setup.

In our earlier discussion, we described DLS as essentially due to microscopic birefringent particles with random orientations, and we neglected the issue of simple macroscopic sample birefringence in our discussion, such as is present in a large anisotropic crystal. Macroscopic birefringence rotates the polarization of a large region of a transmitted beam. Such macroscopic birefringence over a sufficiently large region in a sample would yield a false-positive chirality measurement with use of our device.

We should, however, point out that intended samples for investigation will all be in liquid state, either naturally occurring or made into solutions. The search will be for microscopic life. Thus, we also expect that the samples will be filtered and so be free of macroscopic particles. A homogeneous liquid sample limits the possibility of observing a false positive due to birefringence or the chirality of nonbiological mineral samples, which generally (though not always) occur only for solid materials. This is because any impurities that remain in the filtered liquid sample would exhibit spatially complex, random, linear birefringence on a microscopic scale, *i.e.*, DLS. In our experiments, this was simulated with milk solutions. Detecting chirality in the presence of DLS is the purpose of the polarimeter described in this publication, and we have demonstrated that our polarimeter is considerably more effective than simple crossed polarizers.

Conclusions

Chiral signatures present in extraterrestrial samples provide an excellent indicator for the presence of life in general. However, conventional polarimeters fail in the presence of DLS; and, in an extraterrestrial environment, it is impractical to prepare optically clean samples without DLS. Therefore, it is crucial to be able to make measurements on samples with significant DLS.

The RP polarimeter discussed herein is immune to large amounts of DLS because it measures the *phase* of the desired sinusoidal signal-voltage component of the detected intensity, which is not affected by a significant loss of *amplitude* due to DLS. Consequentially, it provides a significant advantage over other polarimeters for the detection of chirality in the presence of up to 3 orders of magnitude more depolarizing scattering, perhaps more.

On the other hand, the fact that we were able to use a life by-product, milk (which contains lactose, a chiral substance), as our scatterer and yet neglect its chirality in these measurements reflects the fact that the sensitivity of our current polarimeter setup is considerably less than that of a conventional polarimeter operating in the absence of DLS. However, this is an improper comparison. Detecting chirality in the presence of significant DLS is a much more difficult problem than doing so in its absence, which is similar to the problem of imaging through turbid media, another unsolved difficult optical problem. Indeed, for astrobiology, the detection of anything in the absence of DLS is utterly irrelevant, while the problem of detecting chirality in the presence of significant DLS is crucial.

Upon improving the mechanical stability and electronics, we expect to increase the sensitivity of this polarimeter in the presence of DLS to that of conventional polarimeters in the absence of DLS. We are also currently improving this polarimeter design's robustness, compactness, weight, and energy efficiency. We believe that this device could eventually prove space worthy.

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Abbreviations

DLS, depolarizing light scattering; HWP, half-wave plate; QWP, quarter-wave plate; RP, rotating polarization.

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