Summary

Terrestrial biochemistry largely functions through chiral molecules, which determine the functioning and structure of biological systems. Chiral molecules in their most simple form exist in a left-handed (L-) and a right-handed (D-) version, called enantiomers. Importantly, chirality is not an exception but becomes the rule once molecules increase in size. In non-biological systems, a mixture of such molecules is expected to be racemic (50 % - 50 %). A distinctive and characteristic feature of life itself is that these molecules only occur in one configuration: in organisms amino acids occur almost exclusively in the L-configuration and sugars in the D-configuration (homochirality).

The phenomenom of chirality causes a specific response to light. Both the differential absorbance of circularly polarized light and the induced fractional circular polarization of one enantiomer has the exact opposite wavelength dependent spectral shape as that of the other enantiomer. Consequently, in mixtures of abiotic (racemic) molecules these spectra even out to zero, whereas in biotic matter (homochiral) there is a net difference in polarization. These signals can thus be sensed remotely.

As homochirality seems to be exclusive to life (evidence to the contrary is limited to slight chiral excesses on some meteorites, and to molecules that have an origin in biological systems) circular polarization can be used to directly indicate the presence of life beyond Earth and as such is a promising biosignature. Vegetation has an extensive coverage on our planet and is the only life visible on its surface. Consequently, I have extensively studied the polarimetric response of photosynthetic organisms as a proxy for the remote detection of extraterrestrial life.

In **Chapter 1** we give a brief general introduction to this study and we introduce the reader to astrobiology, biosignatures and their remote detection.

These subjects will be covered much more extensively in **Chapter 2**. In this chapter we will give a concise overview of homochirality in life as we know it and introduce recent discussions on the origin of life

and homochirality. Hereafter we explore the interaction between chiral molecules and polarized light and we review the most important mechanisms contributing to the optical phenomena displayed by homochiral molecules. Finally, we will discuss the polarimetry and wavelength considerations for the remote sensing of homochirality and we will point out current and future instrumental possibilities and constraints.

The circular spectropolarimetric instrument 'TreePol' is presented in **Chapter 3**. TreePol was designed for highly sensitive polarimetric measurements both in the laboratory as in-the-field. In the same chapter we present data on the chiroptical evolution of decaying leaves, which showed a remarkable decrease in polarimetric signal while the pigment concentrations remained the same. We attribute this decrease to the deterioration of the chiral macro-aggregates. The dependency of the polarimetric signals on vegetation physiology establishes the robustness as a biosignature, in the sense of a signature of the living state. Additionally, these measurements provide better insight into the possibilities for the monitoring of plant health on Earth.

Leaves have a complex and heterogeneous structure. In addition to the molecular signals created by the photosynthetic machinery, the cell structure and its arrangement within a leaf can create and modify polarization signals. Such intricate phenomena can best be unveiled using complete Mueller matrix imaging polarimetry as the complete Mueller matrix provides the most comprehensive description of the polarimetric responses of a sample. We have investigated this in **Chapter 4**, where we show linear and circular polarization measurements of maple and maize leaves and discuss the corresponding Mueller matrices and the Mueller matrix decompositions, which show distinct features in diattenuation, polarizance, retardance and depolarization. Interestingly, while normal leaf tissue shows a typical split V/I signal with positive and negative bands, the signals close to the veins only display a negative band, a phenomenon possibly caused by the orientations of the chloroplasts.

Algae are an important component of global photosynthesis and are the evolutionairy predecessors of vegetation. Although among most plant species little variation in circular polarizance can be observed, the spectra of algae show a remarkable variation in spectropolarimetric characteristics. Possible, this might be due to differences in the algal photosynthetic structures. In **Chapter 5** we present circular spectropolarimetric measurements of various multicellular algae representing their main groups (green, red and brown algae) and compare them to similar measurements performed on higher plants. While the chloroplasts of these algae are more primitive than those

of higher plants, in certain algae the circular polarizance was much larger; up to 2% as compared to only 1 % in most plants.

Having shown that high quality circular polarimetric spectra can be quickly obtained in the laboratory, the next step is to take dedicated polarimeters into the field. Importantly, compared to the laboratory these instruments will have to cope with dynamic scenes involving, for instance, large changes in illumination and moving targets. Additionally, large amounts of linearly polarized light may be created $> 10\%$ and mitigation of this is crucial. In **Chapter 6** we have investigated the circular polarizance of vegetation in the field and show that life can readily be distinguished from abiotic matter under these complex conditions.

In the general discussion **Chapter 7** we address the findings of the research in this dissertation in a broader context. The fractional circular polarization of light by photosynthetic organisms is a promising biosignature for the detection of extraterrestrial life. Additionally, its use is promising for the remote monitoring of life on Earth. In both cases, the actual application remains challenging. The research in this dissertation paves the way towards these ends, however, much more research is required. Recommendations for future research are provided in this chapter.