Polarimetric investigation of selected cloud compositions in exoplanetary atmospheres

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Received 25 October 2021 / Accepted 29 April 2022

ABSTRACT

Aims. We investigated the impact of selected cloud condensates in exoplanetary atmospheres on the polarization of scattered stellar radiation.

Methods. We considered a selection of 25 cloud condensates that are expected to be present in extrasolar planetary atmospheres. Using the three-dimensional Monte Carlo radiative transfer code POLARIS and assuming Mie scattering theory, we calculated and studied the net polarization of scattered radiation as a function of planetary phase angle at optical to near-infrared wavelengths (0.3 μm to 1 μm).

Results. In addition to the well-known characteristics in the state of polarization, such as the rainbow determined by the real part of the refractive index, the behavior of the underlying imaginary part of the refractive index causes an increase or decrease in the degree of polarization and a change of sign in the polarization at a characteristic wavelength. In contrast to Al₂O₃ and MgFeSiO₃, clouds composed of SiO, MnS, Na₂S, or ZnS produce a rapidly decreasing degree of polarization with increasing wavelength in the context of an exoplanetary atmosphere. Furthermore, the sign of the polarization changes at a wavelength of about 0.5 μm to 0.6 μm, depending on the specific cloud condensate. The resulting net polarization is mainly positive for cloud compositions with large imaginary parts of the refractive index, such as Fe, FeS, and FeO. In addition, for Fe and FeS clouds, the maximum degree of polarization at long wavelengths is shifted to larger phase angles than for FeO.

Conclusions. We found that most of these cloud condensates, such as chlorides, sulfides, or silicates, are distinguishable from each other due to their unique wavelength-dependent complex refractive index. In particular, an increase or decrease of the net polarization as a function of wavelength and a change of sign in the polarization at specific wavelengths are important features for characterizing cloud compositions in exoplanetary atmospheres.

Key words. radiation transfer – methods: numerical – polarization – scattering – planets and satellites: atmospheres

1. Introduction

Clouds are an essential part of atmospheres of most planets in the Solar System, such as sulfuric acid clouds on Venus (Young 1973), water clouds on Earth, or ammonia, ammonium hydrosulfide, and water-ice clouds on Jupiter or Saturn (Atreya et al. 1999). Cloud particles can form and grow when supersaturation occurs. If the saturation vapor pressure curve of a cloud species crosses the planetary pressure-temperature profile, the particles condensate and form clouds (Sánchez-Lavega et al. 2004). Condensation of particles includes simple phase changes (e.g., water, ammonia) or thermochemical reactions, for example, in the case of ammonium hydrosulfide in the atmosphere of Jupiter (Lodders & Fegley 2002). In addition to Solar System objects, the existence of clouds is predicted (e.g., Burrows et al. 1997; Ackerman & Marley 2001; Helling et al. 2001; Lodders & Fegley 2002; Morley et al. 2012). Their signature has been observed in the atmosphere of brown dwarfs and extrasolar planets (e.g., Charbonneau et al. 2002; Pont et al. 2008; Sing et al. 2009; Demory et al. 2011; Gibson et al. 2013; Morley et al. 2013).

Cloud compositions do not only leave an imprint on the transmission spectra from the optical to infrared (Morley et al. 2014; Wakeford & Sing 2015), they also affect the planetary albedo and therefore the observed reflected flux as a function of the planetary phase angle (e.g., Seager et al. 2000). Furthermore, radiation scattered in the planetary atmosphere is usually polarized, whereas the net polarization of inactive solar-type stars is assumed to be negligible (Kemp et al. 1987; Cotton et al. 2017). Consequently, measuring the scattered polarized flux of a planet at various phase angles and wavelengths allows determining atmospheric properties. Hansen & Hovenier (1974) demonstrated this method by characterizing the size distribution and real part of the refractive index of the cloud particles in the atmosphere of Venus using polarimetric observations by Lyot (1929) and Coffeen & Gehrels (1969). Thus, polarimetry also represents a promising tool for the characterization of exoplanetary atmospheres.

With modern instruments, such as HIPPI (Bailey et al. 2015), HIPPI-2 (Bailey et al. 2020), POLISH2 (Wiktorowicz & Nofi 2015), POLLUX (Muslimov et al. 2018), or SPHERE (Beuzit et al. 2019), exoplanet polarimetry has come into reach. Various studies of scattered polarized radiation of exoplanets have been made. Selected examples are predictive studies of the reflected flux and polarization of close-in extrasolar giant planets (Seager et al. 2000), cloud-free Rayleigh scattering planetary atmospheres (Buenzli & Schmid 2009), Jupiter-like gas giants with homogeneous (Stam et al. 2004) and inhomogeneous cloud coverage (Karakildi et al. 2013), Earth-like exoplanets with homogeneous (Stam 2008; Karakildi et al. 2011) and inhomogeneous cloud coverage (Karakildi & Stam 2012), the retrieval of cloud
coverage of Earth-like planets (Rossi & Stam 2017), and the polarization of self-luminous exoplanets (Stolker et al. 2017).

The first detection of polarized scattered radiation of an exoplanet was reported by Berdyugina et al. (2008). The authors were able to obtain polarimetric measurements of the HD 189733 system in B band. The system harbors a planet with an orbital semimajor axis of only about 0.03 AU and a period of about 2.2 days (Bouchy et al. 2005). Assuming Rayleigh scattering, the authors concluded that the observed modulation was due to scattered radiation from the atmosphere of HD 189733b. This first detection was also confirmed by Berdyugina et al. (2011). New observations of HD 189733 by Waktorowicz & Noři (2015) and Bott et al. (2016) also reported a linear polarization signal and confirmed the previously reported observations, but with a lower amplitude of polarization variations. Bailey et al. (2021), however, assumed that the highly variable polarization has its origin in the magnetic activity of the host star.

Another system for which linear polarization was reported by Bott et al. (2018) is WASP-18. This system harbors a planet with an orbital distance of about 0.02 AU and a period of 0.94 days (Hellier et al. 2009). Although the polarization is dominated by the additional influence of the interstellar medium, the authors set an upper limit of 40 ppm on the amplitude of reflected polarized radiation from WASP-18b.

Our study is organized as follows: in Sect. 2 we start with a brief overview of the applied methods. Our model of an exoplanetary atmosphere is described in Sect. 3. Subsequently, the first goal of this study was to review characteristic properties of the polarization state of the radiation that is scattered once (hereafter: single-scattered) by cloud particles. For this purpose, in Sect. 4.1, we calculate optical properties of selected cloud particle compositions that are expected to condensate in various types of planetary atmospheres, ranging from ice giants to hot Jupiters. Our second goal was to investigate the scattered polarized flux of cloudy planetary atmospheres with various cloud compositions, which we report in Sect. 4.2. Therein, we compare the considered cloud compositions, discuss the polarization, and evaluate the feasibility of using characteristic features in the net polarization to distinguish between these cloud particles. To study the polarization at various phase angles and wavelengths ranging from 0.3 μm to 1 μm, the publicly available1 three-dimensional Monte Carlo radiative transfer code POLARIS (Reissl et al. 2016) was applied. It is well tested and has been used to investigate a broad range of astrophysical models such as molecular clouds (Reissl et al. 2017; Pellegrini et al. 2020; Seifried et al. 2020), Bok globules (Brauer et al. 2016; Zielinski et al. 2021), and protoplanetary disks (Heesse et al. 2020; Brunngräber & Wolf 2020, 2021). For the purpose of our study, it has been optimized to handle the radiative transfer in planetary atmospheres (Lietzow et al. 2021). Finally, a discussion and a brief study of the observability of these polarization features is given in Sect. 5. Our conclusions are summarized in Sect. 6.

2. Methods
To calculate the scattered (polarized) flux of an exoplanet, we simulated the radiative transfer in its atmosphere with a Monte Carlo method. We used the three-dimensional Monte Carlo radiative transfer code POLARIS (Reissl et al. 2016), which has recently been optimized to calculate the scattered radiation of planetary atmospheres (Lietzow et al. 2021). Therein, the radiation field is represented by photon packages that are emitted by a spatially extended radiation source, propagate through the three-dimensional model space, are scattered by atmospheric particles, and are finally detected by an observer.

In the past, radiative transfer calculations applied to planetary atmospheres with the Monte Carlo approach have also been performed, for instance, by Buentzli & Schmid (2009), García Muñoz & Mills (2015), Emde et al. (2016), Stolker et al. (2017), or Bailey et al. (2018), while Stam et al. (2006) or Rossi et al. (2018), for example, performed calculations based on the adding-doubling method (de Haan et al. 1987).

To describe the state and degree of polarization of the photon packages, we used the Stokes formalism (e.g., Bohren & Huffman 1983). Here, each photon package carries a wavelength-dependent Stokes vector \( \mathbf{S} = (I, Q, U, V) \). Its four components are the total flux \( I \), the components \( Q \) and \( U \), describing the linear polarization, and the circularly polarized flux \( V \). At each interaction of the photon package with an atmospheric particle, the Stokes vector is multiplied by a scattering matrix \( \mathbf{F}(\theta, \phi) \) to account for the change in polarization,

\[
\mathbf{S}' = \mathbf{F}(\theta, \phi)\mathbf{S},
\]

where \( \mathbf{S}' \) is the Stokes vector of the incoming radiation, and \( \theta \) and \( \phi \) are the scattering angles. When the photon package leaves the atmosphere, the resulting Stokes vector can be measured by an observer. The degree \( P_1 \) and angle \( \chi \) of linear polarization is

\[
P_1 = \frac{\sqrt{Q^2 + U^2}}{I}, \quad \tan(2\chi) = \frac{U}{Q}.
\]

In studies of Solar System objects, a common approach is to express the degree of polarization in a reference system. In particular, the degree of polarization is defined as

\[
P_s = P_1 \cos(2\theta_t), \quad \theta_t = \chi - \left(\psi \pm \frac{\pi}{2}\right),
\]

where \( \psi \) is the position angle of the scattering plane, that is, a plane containing star, planet (or comet), and observer. The term inside the brackets must satisfy the condition \( 0 \leq (\psi + \pi/2) \leq \pi \) (Chernova et al. 1993). As a consequence, the degree of polarization has a sign, and we can distinguish between positive and negative polarization, which means that the radiation is polarized perpendicular and parallel to the scattering plane, respectively. Furthermore, if the planet is symmetric with respect to the scattering plane, it is \( U = 0 \), and with \( \psi = 0 \), we define a signed degree of linear polarization such as

\[
P_s = -\frac{Q}{I}
\]

In this study, the total flux only contains the net scattered planetary flux and not the stellar contribution. The planet can therefore be spatially resolved from its star.

In the case of a homogeneous spherical particle, the scattering matrix given in Eq. (1) is reduced to

\[
\mathbf{F} = \begin{bmatrix}
F_{11} & F_{12} & 0 & 0 \\
F_{21} & F_{22} & 0 & 0 \\
0 & 0 & F_{33} & F_{34} \\
0 & 0 & -F_{34} & F_{33}
\end{bmatrix},
\]

The four essential elements depend on the complex refractive index \( n_c = n_r + i n_i \) of the particle and the size parameter \( x = 2\pi r/\lambda \), where \( r/\lambda \) is the ratio of particle radius and wavelength.

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1 https://portia.astrophysik.uni-kiel.de/polaris/
For a collection of particles with a size distribution \( n(r) \), we define an averaged matrix element as

\[
\langle F_{ij} \rangle = \frac{\int_{r_{\text{min}}}^{r_{\text{max}}} n(r) F_{ij}(r) \, dr}{\int_{r_{\text{min}}}^{r_{\text{max}}} n(r) \, dr}.
\]

(6)

Considering incident unpolarized radiation, the degree of polarization of single-scattered radiation by a collection of particles following Eqs. (1) and (4) becomes

\[
P_s = \frac{\langle F_{22} \rangle}{\langle F_{11} \rangle}.
\]

(7)

3. Model setup

We used a spherical model space that contains the planet and the illuminating source (star). The planet is located at the center of coordinate space, and the star is located at a distance \( d \) from the planet. The planetary atmosphere was divided into a logarithmically spaced radial pressure grid, ranging from a top pressure of \( 10^{-5} \) bar to a pressure of 100 bar. The atmosphere consisted of molecular hydrogen. The total scattering optical depth of the gaseous layers was approximately 18 at 0.5 \( \mu \text{m} \).

Assuming an absorbing surface underneath, the lower boundary represents a thick cloud layer that absorbs all incoming radiation. However, the impact of the lower boundary of the atmosphere is negligible, especially at shorter wavelengths, since the information of the polarization state is lost at this optical depth (Buenzli & Schmid 2009). For the gaseous particles, we only considered scattering and ignored absorption (see Sect. 5 for further discussion). The scattering cross-section, the refractive index, and the scattering matrix were calculated using the formula given by Sneep & Ubachs (2005), Cox (2000), and Hansen & Travis (1974), respectively. For molecular hydrogen, a depolarization factor of 0.02 was used (Hansen & Travis 1974).

In addition to the gaseous layers, we also considered clouds in the atmosphere. The clouds consist of small spherical particles whose distribution is characterized by a power law with exponential decay, such as

\[
n(r) \propto r^{p_1} e^{-r/r^{p_2}},
\]

(8)

where \( r \) is the particle radius. Because of its simplicity, it is commonly applied to describe various types of clouds (Hansen 1971). This size distribution is sometimes also referred to as the standard (gamma) distribution (Hansen & Travis 1974). The two parameters \( p_1 \) and \( p_2 \) depend on the effective radius \( r_{\text{eff}} \) and effective variance \( \nu_{\text{eff}} \) of the distribution

\[
p_1 = 1 - 3\nu_{\text{eff}}/r_{\text{eff}}, \quad p_2 = r_{\text{eff}}^3\nu_{\text{eff}}.
\]

(9)

In contrast to the gaseous particles, we considered both scattering and absorption by the cloud particles. The absorption and scattering cross-section as well as the scattering matrix of the cloud particles were determined using the program mieX (Wolf & Voshchinnikov 2004), which is based on the Mie scattering theory (Mie 1908). An underlying assumption of this approach is that the particles are chemically homogeneous spheres.

Although cloud condensates in planetary atmospheres, such as water droplets, water-ice particles or solid particles in general, can be expected not to be perfectly spherical, the assumption of spherical particles can be used to mimic optical properties, such as cross-sections, of nonspherical particles (Kitzmann et al. 2010). However, polarization is much more sensitive to the particle shape. Linear polarization of randomly oriented nonspherical particles strongly depends on the aspect ratio (Mishchenko & Travis 1994). In addition, for size parameters \( \gtrsim 5 \) and large scattering angles, calculations assuming spheres usually do not agree with the measurements of nonspherical particles, but rounded particles are closely approximated by the Mie scattering theory (Perry et al. 1978). Furthermore, while liquid cloud condensates can be approximated by spheres, such as water clouds on Earth (Goloub et al. 1974) or sulfuric acid clouds on Venus (Hansen & Hovenier 1974; García Muñoz et al. 2014), solid particles, such as ice crystals in cirrus clouds, show different polarization signals compared to liquid water clouds due to their large variability in shape, size, and density (Hess 1998; Goloub et al. 2000). These ice particles in cirrus clouds, however, which were observed to be columns, rosettes, and bullet rosettes in shape, are usually several hundreds of micrometers in size, while smaller micrometer-sized particles appear to be spherical (Lawson et al. 1998).

For this study, we therefore assumed small spherical particles with \( p_1 = 7 \) and \( p_2 = 0.1 \mu \text{m} \), defining the size parameter distribution to apply the Mie scattering theory. These parameters correspond to an effective particle radius of 1 \( \mu \text{m} \) and an effective variance of 0.1 (see Eq. (9)). Probing larger and irregularly shaped particles, such as water-ice particles (see Karalidi et al. 2012), was beyond the scope of this paper.

The cloud layer was located between 1 bar and 0.1 bar. The choice of the size parameter and pressure values was based on previously motivated assumptions of micrometer-sized particles at pressures of several hundred millibar in cloudy atmospheres, for example, in the atmosphere of Jupiter (West et al. 1986) and Venus (Hansen & Hovenier 1974; Knibbe et al. 1997). The parameter values for the standard size distribution are also in agreement with the cloud model by Stam et al. (2004) for a Jupiter-like planet. In addition, in the case of HD 189733b, three-dimensional radiative-hydrodynamics with kinetic microphysical mineral cloud formation processes show that mean cloud particle sizes are typically submicron to micron at pressures lower than 1 bar (Lee et al. 2016).

Pérez-Hoyos et al. (2005) presented a study of the vertical structure of clouds in the atmosphere of Saturn and found a strong latitudinal dependence on its optical thickness in the visible wavelength region, ranging from 20 to 45 at the equator, but 5 at the pole. Since the main goal of this study was to investigate the influence of different cloud compositions, and to avoid introducing too many variables, our planetary model had a homogenous cloud cover. The vertical optical depth of the cloud layer was chosen to be equal to 10 at 0.5 \( \mu \text{m} \), which falls in the range assumed for the clouds of Saturn (Pérez-Hoyos et al. 2005), and also represents the approximate visible mean optical depth of the clouds of Jupiter as proposed by Sato & Hansen (1979).

Depending on the temperature profile in the atmosphere and its composition, clouds can condensate in various pressure regions (e.g., Lodders & Fegley 2002; Visscher et al. 2006, 2010), with condensates grown up to several hundreds of micrometers in size (Hellings et al. 2008; Lee et al. 2015). However, investigating the latitudinal and longitudinal dependence on the height and optical depth of clouds as well as the dependence on the particle size and its influence on the reflected polarized flux were beyond the scope of the current study. Therefore, the planetary model discussed above served as a reference model for all cloud types, regardless of the composition.

The compositions considered here (see Table 1) are expected to condensate in different types of atmospheres of extrasolar
Table 1. Different cloud condensates, and the reference for the complex refractive index.

<table>
<thead>
<tr>
<th>Formula</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al₂O₃</td>
<td>Koike et al. (1995)</td>
</tr>
<tr>
<td>C</td>
<td>Draine (2003)</td>
</tr>
<tr>
<td>CaTiO₃</td>
<td>Ueda et al. (1998)</td>
</tr>
<tr>
<td>CH₄</td>
<td>Martonchik &amp; Orton (1994)</td>
</tr>
<tr>
<td>Cr</td>
<td>Lynch &amp; Hunter in Palik (1991)</td>
</tr>
<tr>
<td>Fe</td>
<td>Lynch &amp; Hunter in Palik (1991)</td>
</tr>
<tr>
<td>FeO</td>
<td>Henning et al. (1995)</td>
</tr>
<tr>
<td>FeS</td>
<td>Pollack et al. (1994)</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>A.H.M.J. Triaud, unpublished</td>
</tr>
<tr>
<td>Fe₂SiO₄</td>
<td>Fabian et al. (2001)</td>
</tr>
<tr>
<td>H₂O</td>
<td>Warren &amp; Brandt (2008)</td>
</tr>
<tr>
<td>KCl</td>
<td>Palik in Palik (1985)</td>
</tr>
<tr>
<td>MgAl₂O₄</td>
<td>Tropf &amp; Thomas in Palik (1991)</td>
</tr>
<tr>
<td>MgFeSiO₄</td>
<td>Dorschner et al. (1995)</td>
</tr>
<tr>
<td>MgO</td>
<td>Roessler &amp; Huffman in Palik (1991)</td>
</tr>
<tr>
<td>Mg₂SiO₃</td>
<td>Jäger et al. (2003)</td>
</tr>
<tr>
<td>MnS</td>
<td>Huffman &amp; Wild (1967)</td>
</tr>
<tr>
<td>NaCl</td>
<td>Eldridge &amp; Palik in Palik (1985)</td>
</tr>
<tr>
<td>Na₂S</td>
<td>Khachai et al. (2009)</td>
</tr>
<tr>
<td>NH₃</td>
<td>Martonchik et al. (1984)</td>
</tr>
<tr>
<td>SiO</td>
<td>Philipp in Palik (1985)</td>
</tr>
<tr>
<td>SiO₂</td>
<td>Philipp in Palik (1985)</td>
</tr>
<tr>
<td>TiO₂</td>
<td>Zeidler et al. (2011); Siefke et al. (2016)</td>
</tr>
<tr>
<td>ZnS</td>
<td>Palik &amp; Addamiano in Palik (1985)</td>
</tr>
</tbody>
</table>

Notes. Most of the data is provided by Kitzmann & Heng (2018) as part of the open-source Exoclime Simulation Platform. "Database of Optical Constants for Cosmic Dust, Laboratory Astrophysics Group of the AIU Jena.

planets and brown dwarfs (Kitzmann & Heng 2018). Methane (CH₄), ammonia (NH₃), or water (H₂O) condensate at temperatures of about 80 K to ~350 K (Carlson et al. 1988; Lodders & Fegley 2002). Sulfide or chloride clouds, such as zinc sulfide (ZnS), sylvite (KCl), or sodium sulfide (Na₂S), condensate in the range of about 700 K to ~1100 K and are therefore expected in atmospheres of Y and T dwarfs (Morley et al. 2012). Iron, corundum (Al₂O₃), or silicate clouds, such as enstatite (MgSiO₃) or forsterite (Mg₂SiO₄), usually condensate at high temperatures (~1300 K) in atmospheres of L dwarfs and deep in the atmospheres of T dwarfs (Lodders 2003; Visscher et al. 2010).

Clouds can condensate in either liquid or solid phase. In the case of iron and silicates, liquid clouds are usually found at much higher temperatures and pressures. For example, liquid iron forms at ≥1800 K, liquid enstatite at ≥1851 K, and liquid forsterite at ≥2163 K (Lodders 1999). The liquid condensates may also dissolve other elements and have complex compositions (Lodders 1999). In addition, optical properties of these iron or silicates compositions exist mainly for the solid phase in the literature.

When we compare the refractive index of the solid and liquid states of CH₄, H₂O, or NH₃, for instance, measurements show a difference in the real part of the refractive index of about 0.05 at 0.5 μm for CH₄ (Martonchik & Orton 1994, interpolated), 0.03 at 0.5 μm for H₂O (Hale & Query 1973; Warren & Brandt 2008), or 0.06 for the sodium D line for NH₃ (Marcoux 1969). Although these differences in the real part of the refractive index are minimal, they have an impact on the resulting optical properties, such as cross-sections and the scattering matrix. For H₂O, the reflected radiation of Earth-like exoplanets with liquid and solid water clouds was studied, for example, by Karalidi et al. (2012). In this study, we assumed Uranus- and Neptune-like or Jupiter-like cloud compositions, that is, methane-ice clouds (Lindal et al. 1987, 1990; Lindal 1992) or ammonia-ice and water-ice clouds (Atreya et al. 1999) are expected, respectively. Furthermore, Morley et al. (2014) modeled atmospheres of Y dwarfs and exoplanets and found that water condenses in the upper atmosphere to form ice clouds. Thus, we investigated the case of solid cloud condensates in the atmosphere.

4. Results

Our goal was to determine the feasibility of distinguishing various cloud condensates expected in exoplanetary atmospheres using polarimetric observations. Therefore, we first reviewed the characteristic properties of the polarization resulting from single scattering by individual cloud particles. Subsequently, we investigated the scattered radiation of a three-dimensional planetary atmosphere consisting of gas and cloud layers as described in Sect. 3, taking multiple scattering events as well as absorption of the cloud particles and the ground layer into account.

4.1. Polarization characteristics of individual particles

As a preparatory study for the investigation of the polarization due to scattering in exoplanetary clouds, we analyzed and reviewed characteristic features that can be found in the polarization due to single scattering by potential cloud particles (Hansen & Travis 1974; Bailey 2007; Karalidi et al. 2011). For this purpose, we calculated the corresponding scattering matrix, based on the wavelength-dependent complex refractive index of the various condensates (see Table 1). Figure 1 shows the degree of polarization given by Eq. (7) for single-scattered radiation as a function of wavelength and scattering angle for various compositions. In general, materials with similar refractive indices also show a similar polarization pattern. Furthermore, materials with a high imaginary part of the refractive index (≥10⁻³) mainly show a positive polarization, while materials with a low imaginary part of the refractive index (≤10⁻³) also show a negative polarization.

For H₂O and CH₄ (see Fig. 1) with a real part of the refractive index of about 1.31 and 1.32, respectively, at a wavelength of 0.5 μm, there is a broad plateau of positive polarization at a scattering angle of approximately 150°. This is the result of rays that are reflected inside the particle once, also called the primary rainbow. This has been discussed, for example, by Liou & Hansen (1971), Hansen & Travis (1974), or Bailey (2007). The plateau of positive polarization ranges over the entire considered wavelength range. For scattering angles below 90°, the degree of polarization mainly has a negative sign, which is a consequence of rays that are refracted twice at the particle (Hansen & Travis 1974). At scattering angles of about 20°, there is a second plateau of positive polarization. This is a result of rays that are externally reflected once at the particle (Hansen & Travis 1974). This plateau ranges up to a wavelength of about 0.5 μm.

For increasing wavelengths, the rainbow is shifted toward shorter wavelengths and larger scattering angles (e.g., Liou & Hansen 1971; Hansen & Travis 1974; Bailey 2007). This is the case for NH₃ with a real part of the refractive index of about
Wavelength $[\mu m]$

Fig. 1. Degree of signed polarization $P_1$ of single-scattered radiation (see Eq. (7)) for various cloud types as a function of wavelength and scattering angle ($\theta = 0^\circ$; forward scattering). In addition, the approximate value of the complex refractive index at a wavelength of $0.5 \, \mu m$ is shown in the top left corner of each figure. See Sect. 4.1 for details.

1.44 at a wavelength of $0.5 \, \mu m$, or for KCl with a real part of the refractive index of about $1.5$ at $0.5 \, \mu m$. For increasing wavelengths, a plateau of negative polarization forms at large wavelengths, as also discussed by Hansen & Travis (1974). In addition, at small scattering angles of about $5^\circ$, the plateau of positive polarization ranges up to a wavelength of about $0.6 \, \mu m$ for NH$_3$ and $0.7 \, \mu m$ for KCl. Similar features were discussed by Hansen & Hovenier (1974) in the context of the analysis of scattered light polarimetry of Venus. The real part of the refractive index of sulfuric acid clouds in the atmosphere of Venus is about 1.44 and is therefore similar to the real refractive index of ammonia.

This shift of the rainbow with increasing real part of the refractive index is valid up to a refractive index of about 1.547 (Nussenzveig 1969). Hence, for compositions such as MgAl$_2$O$_4$, MgO, or Fe$_2$SiO$_4$, the positive polarization shifts back to longer wavelengths, as also discussed by Hansen & Travis (1974). In addition, for a real part of the refractive index of $\geq 1.7$, the plateau of positive polarization at small scattering angles ranges over the entire considered wavelength region. However, a peak of positive polarization at the end of this plateau, which is described by Hansen & Travis (1974) and is a result of anomalous diffraction (van de Hulst 1957), is not visible here.

For a real part of the refractive index $>2$, such as CaTiO$_3$, TiO$_2$, or ZnS, two plateaus of positive polarization form at large
scattering angles. One plateau is located at a scattering angle of about 150° and ranges from a wavelength of 0.4 to ~0.7 μm. The second plateau is located at scattering angles of about 180° and ranges over the entire considered wavelength region.

For an increasing imaginary part of the refractive index, the amount of radiation traveling through the particle decreases. As a consequence, features such as negative polarization due to double refraction, or positive polarization due to internal reflections are lost, and radiation reflected from outside the particle with a positive degree of polarization dominates the polarization pattern (Hansen & Travis 1974). Thereby, the maximum degree of polarization is shifted toward 180°−2θn, where θn is the Brewster angle (Brewster 1815). This is the case for C, Cr, Fe, FeO, FeS, or Fe2O3. Furthermore, for compositions with a very large imaginary part of the refractive index (≥3), such as Fe or Cr, the degree of linear polarization of single-scattered radiation decreases compared to compositions such as FeO or FeS with an imaginary part of the refractive index of ≤1.4 at 0.5 μm. This applies to compositions with a high absolute value of the complex refractive index in general.

For compositions such as Al2O3, MgFeSiO3, MnS, or SiO, there is a high imaginary part of the refractive index at short wavelengths and a small imaginary part of the refractive index at long wavelengths. At short wavelengths, the scattered radiation is therefore dominated by a high degree of positive polarization, while with increasing wavelength, the polarization decreases and becomes negative at a characteristic wavelength.

### 4.2. Polarization of an exoplanetary atmosphere

Although it is possible to characterize the refractive index of the material by its single-scattered polarized radiation, as discussed in Sect. 4.1, multiple scattering and additional Rayleigh scattering by gaseous particles influence the polarized radiation scattered in a cloudy exoplanetary atmosphere (e.g., Karalidi et al. 2011). It therefore remained to be investigated whether the considered cloud compositions with their characteristic polarization features identified in Sect. 4.1 due to, for instance, a wavelength-dependent imaginary part of the refractive index, are distinguishable in this significantly more complex environment. Based on the model described in Sect. 3, we calculated the scattered polarized radiation of a cloudy planetary atmosphere. In Sects. 4.2.1 to 4.2.4, we investigated and compared different cloud compositions depending on the properties of their refractive index.

At shorter wavelengths (~0.3 μm), the degree of polarization for all atmospheric models is in general positive, with a maximum value around a phase angle of 90° due to Rayleigh scattering of the gaseous layers above the clouds, as discussed, for example, by Hansen & Hovenier (1974), Stam (2008), or Karalidi et al. (2011). However, with increasing wavelength, the impact of the gaseous particles weakens because of the strong wavelength dependence on the Rayleigh scattering cross-section (α ∝ λ−4). Therefore, the degree of polarization of the cloud particles starts to dominate the net scattered radiation (Karalidi et al. 2011). As a result of multiple scattering in the atmosphere, the degree of polarization is lower than that of single scattering (Hansen & Travis 1974; Karalidi et al. 2011). However, although multiple scattering decreases the degree of polarization, it does not change the sign of polarization because the reflected radiation is dominated by single-scattered radiation from the upper layers of the clouds (Karalidi et al. 2011). Thus, the features discussed in Sect. 4.1 are still imprinted on the scattered polarized flux. In the following and in contrast to Fig. 1, we describe the scattered polarized flux as a function of the planetary phase angle α. For single-scattered radiation, it is α = 180° − θ.

#### 4.2.1. Clouds with various real parts of the refractive index

In Fig. 2, the signed degree of linear polarization (P) of various model atmospheres is shown as a function of wavelength and phase angle. The model atmospheres have clouds consisting of H2O, NH3, MgSiO3, MgAl2O4, Fe2SiO4, or CaTiO3. These different cloud compositions have a small imaginary part of the refractive index (~10−3 at 0.5 μm) in common and cover a broad range of the real part of the refractive index (~1.34 to ~2.36 at 0.5 μm). In this case, our simulations confirm that the characteristic polarization features due to the real part of the refractive index can be used to distinguish these different materials and to characterize the refractive index. Especially the locations of the positive degree of polarization at small phase angles (or at large scattering angles) have been discussed previously by various authors (e.g., Hansen & Hovenier 1974; Bailey 2007; Karalidi et al. 2011).

For clouds with a real part of the refractive index of about 1.55 to 1.75 such as MgSiO3 and MgAl2O4, the plateau of negative polarization at small phase angles (~0°) is visible for the entire considered wavelength range. In addition, for cloud condensates with a real part of the refractive index of ≥1.75 such as MgAl2O4, Fe2SiO4, and CaTiO3, the plateau of the positive degree of polarization at large phase angles ranges over the entire considered wavelength region as well. For CaTiO3 clouds, there is a small spot of zero polarization at a phase angle of about 15° and at a wavelength of about 0.38 μm, surrounded by a positive degree of polarization. Furthermore, the degree of polarization increases at small wavelengths (~0.3 μm) for CaTiO3 because the imaginary part of the refractive index increases slightly at these wavelengths.

Although Karalidi et al. (2011) considered liquid-water clouds, the results of our simulations for water clouds are in agreement with those obtained by Karalidi et al. (2011) because the authors used similar parameter values for the size distribution and the difference in the refractive index comparing liquid and solid water is small (see Sect. 3). In contrast, in the study by Karalidi et al. (2012), the plateau of positive polarization at phase angles about 30° solely arises from internal reflections of liquid-water clouds and not from water-ice particles because the authors assumed nonspherical ice crystals.

The polarization curves of close-in extrasolar giant planets by Seager et al. (2000) or the polarization curves applied to HD 189733b by Koppura et al. (2016) and Bailey et al. (2018) assumed cloud condensates, such as iron or silicates. Seager et al. (2000) considered a homogeneous MgSiO3-Al2O3-Fe cloud, and Koppura et al. (2016) assumed a silicate cloud with a refractive index of 1.68 + 10−4i. The authors of these two studies observed a peak of polarization at small phase angles, which agrees with our peak of negative polarization at small phase angles. Bailey et al. (2018) compared clouds composed of MgSiO3, Mg2SiO4, Al2O3, or Fe. However, the authors considered particle sizes of r_eff ≤ 0.1 μm, which results in size parameters of ≤1.4. The scattering is therefore almost Rayleigh scattering. Thus, the polarization shows a maximum centered around a phase angle of 90°.

#### 4.2.2. Clouds with a large imaginary part of the refractive index

In Fig. 3, the signed degree of linear polarization (P) is shown for model atmospheres with clouds consisting of FeS, Fe, FeO,
Fig. 2. Degree of signed polarization $P_s$ (see Eq. (4)) as a function of wavelength and phase angle for various cloud compositions. As indicated by the spherical symbols at the top, a phase angle of 0° corresponds to the planet in full phase. See Sect. 4.2.1 for details.

Fig. 3. Same as Fig. 2, but for different cloud compositions. See Sect. 4.2.2 for details.
Fig. 4. Same as Fig. 2, but for different cloud compositions. See Sect. 4.2.3 for details.

Fe$_2$O$_3$, Cr, or C. These different cloud compositions cover a broad range of the real part of the refractive index (~1.36 to ~2.99 at 0.5 µm) and have a large imaginary part of the refractive index (~0.7 at 0.5 µm) in common. Due to the high imaginary part of the refractive index, the single-scattering albedo of the cloud condensates decreases, resulting in a lower impact of multiple scattering and, thus, an increase in the degree of polarization. However, for Fe and Cr clouds, the polarization decreases because the degree of polarization after single scattering decreases (see Sect. 4.1). The maximum value of the degree of polarization is ~0.59 and ~0.47 for Fe and Cr clouds, respectively. For atmospheres composed of FeS, FeO and Fe$_2$O$_3$ clouds, the maximum value of the degree of polarization is ~0.75. In contrast to purely Rayleigh scattering, the maximum degree of polarization is shifted to larger phase angles for these compositions (see Sect. 4.1). At long wavelengths, the maximum degree of polarization is shifted to phase angles of about 125° for FeS, Fe, and Cr clouds, while for Fe$_2$O$_3$ and C clouds, the maximum is shifted to phase angles of about 115°, and to about 110° for FeO clouds. Since the imaginary part of the refractive index decreases at longer wavelengths for Fe$_2$O$_3$, there is a plateau of negative polarization for atmospheres composed of Fe$_2$O$_3$ at phase angles of about 30°.

4.2.3. Clouds with a variable imaginary part of the refractive index

In Fig. 4, the signed degree of linear polarization ($P_\circ$) is shown for model atmospheres with clouds consisting of Al$_2$O$_3$, MgFeSiO$_4$, SiO, MnS, Na$_2$S, or ZnS. These different cloud compositions cover a broad range of the real part of the refractive index (~1.58 to ~2.95 at 0.5 µm) and have a varying imaginary part of the refractive index in the considered wavelength region. For example, for MnS, the imaginary part of the refractive index decreases from about 1.3 at 0.3 µm to about 2.7 × 10$^{-5}$ at 1 µm. In this case, the reflected radiation has solely a high degree of positive polarization at short wavelengths, but characteristic positive as well as negative polarization features at long wavelengths. Thus, at wavelengths ≥0.6 µm, the plateaus of positive and negative polarization determine the real part of the refractive index of the composition. At short wavelengths, the behavior of the positive polarization is linked to the imaginary part of the refractive index. In contrast to Al$_2$O$_3$ and MgFeSiO$_4$ clouds, the positive polarization for an atmosphere composed of SiO, MnS, Na$_2$S, and ZnS clouds decreases rapidly with increasing wavelength. At a characteristic wavelength of about 0.5 to 0.6 µm, depending on the material, the sign of the polarization changes. This is the result of the strong decrease of the imaginary part of the refractive index at these wavelengths. For atmospheres composed of Al$_2$O$_3$ and MgFeSiO$_4$ clouds, no such characteristic wavelength can be defined. Instead, the sign of polarization changes at different wavelengths for different planetary phase angles. In addition, for Al$_2$O$_3$, MgFeSiO$_4$, and Na$_2$S clouds, the degree of negative polarization at long wavelengths is larger than for cloud compositions such as SiO, MnS, or ZnS. This is due to the larger imaginary part of the refractive index of Al$_2$O$_3$, MgFeSiO$_4$, and Na$_2$S at these wavelengths, resulting in a smaller single-scattering albedo and, thus, a lower impact of multiple scattering.

4.2.4. Clouds with an equal real part of the refractive index

In Fig. 5, the signed degree of linear polarization ($P_\circ$) is shown for model atmospheres with clouds consisting of KCl, MgSiO$_3$, NaCl, SiO$_2$, Al$_2$O$_3$, and Mg$_2$SiO$_4$, which have real part of the refractive index of about 1.50 to 1.62 at 0.5 µm. Except for
Al₂O₃, the compositions have a small imaginary part of the refractive index (≤10⁻² at 0.5 μm) as well. The plateau of positive polarization at large phase angles and the plateau of negative polarization at small phase angles show similar characteristics, regardless of the material. The most significant difference is found for an atmosphere consisting of Al₂O₃ clouds. The plateau of positive polarization at large phase angles and at long wavelengths reveals a real part of the refractive index, similar to Mg₂SiO₄ clouds. However, the higher degree of polarization for Al₂O₃ clouds compared to the other compositions indicates a smaller single-scattering albedo and, thus, a larger imaginary part of the refractive index.

Compositions with a similar behavior of real as well as imaginary part of the refractive index in the considered wavelength region, such as MgSiO₃, NaCl, or SiO₂, cannot be distinguished from each other. However, NaCl and MgSiO₃ have different condensation temperatures of ~800 (Burrows & Sharp 1999) and ~1600 K (Visscher et al. 2010), respectively. Thus, NaCl potentially condenses in atmospheres of T dwarfs, for example, while MgSiO₃ will more likely be present in atmospheres of M dwarfs or deep in atmospheres of L dwarfs. In addition, as discussed by Visscher et al. (2010), if MgSiO₃ condenses in the atmosphere, it removes silicon from the gas phase, preventing SiO₂ (~1550 K, Visscher et al. 2010) from condensing.

5. Discussion

In this section, we address selected parameters and corresponding questions that also have an impact on the net polarization, but were not covered in our simulations. The optical depth of the cloud layer in this study had a fixed value of 10 at 0.5 μm. An increasing optical depth of the cloud layer affects the degree of net polarization because the resulting increasing multiple scattering decreases the net degree of polarization (Karalidi et al. 2011; Bailey et al. 2018). On the other hand, a decreasing cloud optical depth would cause the Rayleigh scattering to dominate the net degree of polarization. This is similar to the case of a cloud layer that is located in higher pressure regions because the optical depth of gaseous particles increases above the cloud layer. Information about the polarization of deep cloud layers is lost at optical depths ≥2 of the upper layers (Buenzli & Schmid 2009). For a detailed discussion of cloud top pressure and cloud optical depth, see Karalidi et al. (2011).

The size distribution of the cloud particles is another crucial parameter determining the scattering properties of the particles. While the effective radius causes the degree of polarization to shift along the wavelength axis, increasing the effective variance causes various polarization features to smooth out (Hansen & Hovenier 1974; Hansen & Travis 1974). For smaller particles for a given observing wavelength, that is, smaller size parameters, the scattering is in the Rayleigh regime, resulting in a loss of the characteristic features in the polarization. For a detailed study of various particle sizes, see, for instance, Hansen & Hovenier (1974), Hansen & Travis (1974), Seager et al. (2000), Karalidi et al. (2011, 2012).

In our study, the planetary atmosphere had a homogeneous cloud coverage that covered the entire planet. However, the cloud coverage fraction and an asymmetric coverage of hemispheres have an impact on the scattered flux as well. The influence of the cloud coverage was studied in detail by Karalidi & Stam (2012), and Karalidi et al. (2013), and the retrieval of the cloud coverage of Earth-like exoplanets was studied, for example, by Rossi & Stam (2017). In addition, horizontally inhomogeneous cloud coverage also causes a nonzero circular polarization degree, which was studied by Rossi & Stam (2018).
As already mentioned in Sect. 3, we ignored absorption by gaseous particles. In general, absorption will decrease the effect of multiple scattering, thus, the degree of polarization increases. However, the net degree of polarization can decrease as well if there are, for instance, reflecting clouds in the lower layers. For a detailed discussion and radiative transfer computation with absorption by gaseous particles, see, for example, Stam et al. (2004) or Buenzli & Schmid (2009).

For the degree of polarization shown in Figs. 2–5, we assumed that the exoplanet and its host star are spatially separated observationally, thus, the high fraction of unpolarized stellar radiation is not included in the net polarization. Considering an exoplanet-star system where both components cannot be separated observationally, the ratio of the polarized planetary flux and the total stellar flux, that is, the polarization contrast, is given by (Hunziker et al. 2020)

$$C_{\text{pol}} = \frac{P(\alpha, \lambda)I(\alpha, \lambda)}{d^2}. \quad (10)$$

Here, $P$ and $I$ are the phase angle and wavelength-dependent degree of polarization and reflectivity of the planet, respectively. At a phase angle of 0°, the reflectivity is equivalent to the planetary geometric albedo. Furthermore, $r$ and $d$ are the planetary radius and distance between planet and star, respectively. To calculate the planetary temperature profile, we followed Guillot (2010) and took parameter values based on HD 209458b ($k_{\text{th}} = 10^{-2} \text{ cm}^2 \text{ g}^{-1}$, $k_{\text{e}} = 4 \times 10^{-3} \text{ cm}^2 \text{ g}^{-1}$, $T_{\text{int}} = 500 \text{ K}$, Guillot 2010; $r = 1.45 R_\star$, $T_\star = 6092 \text{ K}$, Boyajian et al. 2015; $m = 0.64 M_\odot$, Snellen et al. 2010). However, we set $d = 1 \text{ AU}$ with an observational distance of 5 pc to be suitable for SPHERE/ZIMPOL (Hunziker et al. 2020). Thus, we obtained temperatures of about 500 to ~1000 K in the pressure region of about 10 mbar to ~1 bar, which means that compositions such as KCl, NaCl, Na$_2$S, or ZnS potentially form cloud condensates.

Figure 6 shows the polarization contrast (Eq. (10)) as a function of wavelength and planetary phase angle for various compositions that are expected to condensate in the atmosphere, and a cloud-free atmosphere for comparison. For the cloud-free atmosphere, the polarization contrast is highest with maximum values of about $3.7 \times 10^{-8}$ at phase angles of about 65° across the considered wavelength region. For the cloudy atmospheres, the polarization contrast is highest at short wavelengths (~0.35 μm) with values of $\sim 2 \times 10^{-8}$ due to Rayleigh scattering. In addition, for NaCl clouds, the polarization contrast reaches values of $\geq 2 \times 10^{-8}$ at small phase angles and long wavelengths, making the plateau of negative polarization detectable, for instance, with SPHERE/ZIMPOL (Hunziker et al. 2020). For Na$_2$S and ZnS clouds, the high degree of polarization at short wavelengths with a rapidly decreasing degree of polarization for increasing wavelength, as discussed in Sect. 4.2.3, produces a polarization contrast of $\leq 10^{-8}$ because the total reflected flux decreases with decreasing single-scattering albedo as well.

6. Conclusions

In the first part of this study (Sect. 4.1), we investigated the single-scattered polarized radiation of various cloud condensates, which are expected in planetary atmospheres. We reviewed characteristic properties of the polarization state depending on the scattering angle, wavelength, and refractive index of the cloud particle. As demonstrated in selected earlier studies, we confirmed the significant impact of the chemical composition...
and, thus, the complex refractive index of the cloud particles on the state of polarization of the scattered radiation.

If the imaginary part of the refractive index is small, refracted rays dominate the polarization pattern (Hansen & Travis 1974). Characteristic features, such as the rainbow, depend on the real part of the refractive index (e.g., Liou & Hansen 1971; Hansen & Travis 1974; Bailey 2007). However, most of the considered cloud condensates have a nonnegligible imaginary part. For an increasing imaginary part of the refractive index, polarization features resulting from rays that are refracted or reflected outside the particle are lost, and radiation that is reflected externally dominates (Hansen & Travis 1974). For compositions with a large imaginary part of the refractive index over the entire considered wavelength region, the polarization is positive, with the maximum value shifted to smaller wavelengths with a large imaginary part of the refractive index, polarization features resulting from rays that are refracted or reflected inside the particle are lost, and radiation that is reflected externally dominates (Hansen & Travis 1974). For compositions with a large imaginary part of the refractive index, polarization features resulting from rays that are refracted or reflected outside the particle are lost, and radiation that is reflected externally dominates (Hansen & Travis 1974). For compositions with a large imaginary part of the refractive index over the entire considered wavelength region, the polarization is positive, with the maximum value shifted to smaller scattering angles (see Hansen & Travis 1974). Compositions such as Al$_2$O$_3$, MgFeSiO$_4$, SiO, MnS, Na$_2$S, and SiO have a strongly varying imaginary part of the refractive index. At short wavelengths, the polarization is therefore mainly positive and becomes negative at longer wavelengths. In between, there is a change of sign of the polarization due to the decrease of the imaginary part. The wavelength at which the sign changes depends on the chemical composition and, thus, allows for the identification of the chemical composition of the respective condensate.

The goal of the second part of this study (Sect. 4.2) was to evaluate the feasibility to distinguish between the different considered cloud materials in a planetary atmosphere. For this purpose, we simulated the reflected polarized flux of cloudy planetary atmospheres with selected cloud compositions investigated in Sect. 4.1. These simulations were performed with the three-dimensional Monte Carlo radiative transfer code POLARIS, which was optimized to handle the radiative transfer in planetary atmospheres. The degree of linear polarization was studied at planetary phase angles ranging from $0^\circ$ to $180^\circ$ at wavelengths from 0.3 $\mu$m to 1 $\mu$m.

Building on the results of the single-scattering polarization obtained in the first part of our study, we first presented results for compositions with a small imaginary, but various real parts of the refractive index in Sect. 4.2.1. In agreement with earlier studies (Hansen & Hovenier 1974), our simulations confirmed that the polarization features such as the rainbow can be used to determine the refractive index of the cloud particles.

For high values of the imaginary part of the refractive index, considered in Sect. 4.2.2, the polarization is mainly positive and the overall degree of polarization increases. However, each of the considered cloud composition shows a characteristic behavior of the polarization, depending on the individual imaginary part of the refractive index.

- Cloud compositions with an imaginary part of $\geq 3$, such as Fe and Cr, show a decrease in the degree of polarization compared to compositions such as FeS and FeO.
- For FeS, Fe, and Cr clouds, the maximum degree of polarization at long wavelengths is shifted to phase angles of about $125^\circ$, while for FeO clouds, the maximum degree of polarization is shifted to phase angles of about $110^\circ$.
- For Fe$_2$O$_3$, there is a plateau of negative polarization at wavelengths of $\geq 0.9 \mu$m because the imaginary part of the refractive index is decreasing rapidly with increasing wavelength.

If the imaginary part of the refractive index strongly varies in the considered wavelength region (Sect. 4.2.3), the behavior of the degree of polarization and a change of the sign of the polarization degree can be linked to the behavior of the imaginary part of the refractive index of the condensate, allowing one to characterize the cloud composition.

- Clouds composed of Al$_2$O$_3$ and MgFeSiO$_4$ show a smooth decrease of positive polarization with increasing wavelength.
- Clouds composed of SiO, MnS, Na$_2$S, and ZnS show a rapidly decreasing degree of polarization with increasing wavelength. In addition, at a characteristic wavelength of about 0.5 to 0.6 $\mu$m, depending on the imaginary part of the refractive index, the sign of the polarization changes.

Finally, for condensates with similar values of the real part of the refractive index (Sect. 4.2.4), a distinction of various compositions is almost impossible.

- The most significant difference in the polarization was identified in the case of Al$_2$O$_3$ clouds. Here, the underlying imaginary part of the refractive index results in a high degree of positive and negative polarization at short and long wavelengths, respectively.
- However, chlorides or sulfides with similar refractive indices as silicates have a different condensation temperature. Thus, compositions with a similar net polarization across the considered wavelength and phase angle region, but different condensation temperatures, can be distinguished indirectly by comparison of the temperatures of the planetary atmosphere and, thus, its potential to host certain condensates/clouds.

In summary, most of the considered compositions that are expected to condensate in exoplanetary atmospheres, such as chlorides, sulfides, or silicates, can be distinguished via polarization measurements due to their unique wavelength-dependent real as well as imaginary part of the refractive index and/or different condensation temperatures.

Acknowledgements. We wish to thank the anonymous referee, for suggestions improving the presentation of the results of this study. This research made use of Astropy (https://www.astropy.org), a community-developed core Python package for Astronomy (Astropy Collaboration 2013, 2018), Matplotlib (https://matplotlib.org) (Hunter 2007), Numpy (https://numpy.org) (Harris et al. 2020), NASA’s Astrophysics Data System (https://ui.adsabs.harvard.edu), and a modified A&A bibliography style file with clickable link in the bibliography.

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