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Martian meteorites reflectance and implications for rover missions

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Highlights

- We provide the first VNIR (0.4–3 μm) spectral database representative of the current Martian meteorites' diversity
- Point measurements and hyperspectral cubes are acquired on unprocessed rock samples, similarly to present and future measurements of the SuperCam and MicrOmega instruments onboard Martian rovers
- Bidirectional spectral measurements are performed on a shergottite rock and powder, in order to characterize the influence of varying illumination and observation geometries on its spectral features

Abstract

During this decade, two rovers will characterize in situ the mineralogy of rocks on Mars, using for the first time near-infrared reflectance spectrometers: SuperCam onboard the Mars 2020 rover and MicrOmega onboard the ExoMars rover, although this technique is predominantly used in orbit for mineralogical investigations. Until successful completion of sample-return missions from Mars, Martian meteorites are currently the only samples of the red planet available for study in terrestrial laboratories and comparison with in situ data. However, the current spectral database available for these samples does not represent their diversity and consists primarily of spectra acquired on finely crushed samples, albeit grain size is known to greatly affect spectral features. Here, we measured the reflected light of a broad Martian meteorite suite as a means to catalogue and characterize their spectra between 0.4 and

40 3 μm . These measurements are achieved using a point spectrometer acquiring data comparable to
41 SuperCam, and an imaging spectrometer producing hyperspectral cubes similarly to MicrOmega. Our
42 results indicate that point spectrometry is sufficient to discriminate the different Martian meteorites
43 families, to identify their primary petrology based on band parameters, and to detect their low content
44 in alteration minerals. However, significant spectral mixing occurs in the point measurements, even at
45 spot sizes down to a few millimeters, and imaging spectroscopy is needed to correctly identify the
46 various mineral phases in the meteorites. Additional bidirectional spectral measurements on a
47 consolidated and powdered shergottite confirm their non-Lambertian behavior, with backward and
48 suspected forward scattering peaks. With changing observation geometry, the main absorption
49 strengths show variations up to $\sim 10\text{--}15\%$. The variation of reflectance levels is reduced for the rock
50 surface compared to the powder. All the spectra presented are provided in the supplementary data for
51 further comparison with in situ and orbital measurements.

52

53 **1 Introduction**

54

55 Reflectance spectroscopy is a non-destructive technique allowing the detection of various
56 mineral phases without the need for sample preparation. Therefore, it is well suited for the
57 characterization of precious samples such as Martian meteorites. It is also the prime technique used to
58 remotely investigate the mineralogical composition of planetary surfaces and has shown great successes
59 in the case of Mars (e.g., Mustard et al., 1993; Erard and Calvin, 1997; Bibring et al., 2005; Ehlmann et al.,
60 2009; Carter et al., 2013). The Infrared Spectrometer of the SuperCam instrument and MicrOmega
61 (respectively a point spectrometer and an imaging spectrometer) are VNIR (Visible and Near-Infrared)
62 reflectance spectrometers used to characterize the mineralogy of the Martian surface. They are operating
63 or will operate onboard the Mars 2020 (NASA; Wiens et al., 2017) and ExoMars rovers (ESA/Roscosmos;
64 Bibring et al., 2017) respectively. These analyses will provide the opportunity to compare and link the
65 spectra measured at the surface to the global spectral measurements from the orbiters, but also to the
66 well-characterized Martian meteorites.

67 Until the Mars Sample Return (MSR) and the Martian Moon eXploration (MMX) campaigns are
68 completed (i.e., 2031 at the earliest for MSR and 2029 for MMX; Usui et al., 2018; Muirhead et al., 2020),
69 Martian meteorites are currently the only material from the Martian system readily available for
70 laboratory studies and direct comparison with in situ and orbital measurements (even though most of
71 them originate from the subsurface and might not be representative of the Martian regolith). However,
72 their documented reflectance spectra were mainly acquired on powders, even though the physical state
73 of the sample, and especially its grain size, have been shown to significantly influence both the absolute
74 reflectance and the shape of the absorption bands (Crown and Pieters, 1987; Salisbury and Wald, 1992;
75 Sunshine et al., 1993; Mustard and Hays, 1997). In addition, in situ measurements by the SuperCam
76 instrument will be achieved remotely and without any sample grinding (Wiens et al., 2017). This
77 demonstrates the need to acquire spectra spanning the diversity of Martian meteorites, especially on
78 non-pulverized rock samples.

79 The documentation of multiple spectra of Martian meteorites would also provide additional
80 constraints on the location of their ejection sites. Assessing these key locations would bring a unique
81 opportunity to link the Martian meteorites extensive characterization in laboratories to the orbital
82 geological study of the surface of Mars. In addition, the comparison between their absolute age and the
83 crater densities of their ejection site would allow the calibration of the Martian chronology, in a joined
84 effort with the sample return missions (e.g., Muirhead et al., 2020). Given their diversity of crystallization
85 and cosmic-ray exposure ages (especially those of shergottites), they probably originate from different
86 sites on Mars (the readers are referred to Nyquist et al., 2001 and Udry and Day, 2018 for a review on
87 their age). As of today, some precise ejection sites have been proposed (e.g., Werner et al., 2014) but
88 there is no real consensus on the source craters of these meteorites. Ody et al. (2015) conducted a global
89 survey of the Martian surface using the Observatoire pour la Minéralogie, l'Eau, les Glaces et l'Activité
90 (OMEGA) dataset and compared the reflectance of the terrains to the spectra of seven Martian
91 meteorites, as a means to locate their ejection sites. This study revealed potential source regions with
92 close spectral matches, but was limited by the number of available meteorite spectra (e.g., only four
93 shergottite spectra, only one nakhlite spectrum, only one chassignite spectrum, and no spectrum of
94 augite-rich basalt or polymict breccia specimen). A new analysis of the orbital global spectral datasets
95 with a more extensive meteorite spectral database might narrow down these potential ejection regions.

96 With the exception of a few studies (e.g., Fadden and Cline, 2005; Hiroi et al., 2011; Manzarini et
97 al., 2019), most of the reflectance spectra previously obtained in the VNIR range on Martian meteorites
98 were acquired on powders (e.g., Sunshine et al., 1993; Bishop et al., 1998; Dyar et al., 2011; Beck et al.,
99 2015, Filiberto et al., 2018). Spectra of the polymict breccia were previously acquired on a chip of NWA
100 7034 and NWA 8171 (Cannon et al., 2015; Izawa et al., 2018) and on a residue of a dry sawing of NWA
101 7533 (Beck et al., 2015). A reflectance survey on rock samples of Martian meteorites in the 0.25-2.5
102 spectral range was conducted by Hiroi et al. (2011) on eight shergottites and four nakhlites, two of which
103 are in common with our sample suite. Similar measurements were performed by Fadden and Cline (2005)
104 as a means to produce a framework to locate ejection regions on Mars. These studies show that VNIR
105 reflectance can successfully be used to characterize the main mineralogy of Martian rocks and classify
106 them relative to the families previously presented. A few additional spectra of Martian meteorites are
107 available in the RELAB spectral library, mostly acquired on powders.

108 In this study, we aim at contributing to a broad Martian meteorites spectral database, by
109 providing their spectra in the VNIR range between 0.4 and 3 μm , using instruments similar to the
110 SuperCam and MicrOmega spectrometers. We use these analyses to characterize their spectral features
111 in the VNIR range. Our spectral survey is conducted on a suite of samples representative of the current
112 Martian meteorites' diversity, the majority of them being rock chips or cut sections. We present two
113 types of VNIR spectral analyses: point spectrometry and imaging spectrometry. These techniques are
114 closely analogous to what is and will be achieved by the reflectance spectrometers in SuperCam
115 (acquiring point spectra) and MicrOmega (acquiring spectral cubes). In addition to the spectra of 11

116 Martian meteorites which were already measured in the literature mostly on particulate samples (ALH¹
117 77005, ALH 84001, EETA² 79001, Lafayette, Los Angeles, MIL³ 03346, Nakhla, NWA⁴ 2737, NWA 7034,
118 Tissint and Zagami), we document the reflectance spectra of 16 meteorites whose VNIR spectra have
119 not been reported to date: NWA 480, NWA 4766, NWA 12633, NWA 12960, NWA12965 (basaltic
120 shergottites), DaG⁵ 476, DaG 489, NWA 1195 (olivine-orthopyroxene-phyric shergottites), NWA 1068,
121 SaU⁶ 008 (olivine-phyric shergottites), NWA 1950, NWA 4468, NWA 7397 (poikilitic shergottites), CeC⁷
122 022, NWA 817 (nakhlites) and NWA 8159 (augite-rich basalt)⁸. Eleven meteorites from our suite,
123 including the polymict breccia NWA 7034, are analyzed with imaging spectroscopy, providing refined
124 details on the spectral features of the different Martian meteorite families and further interpretation of
125 the point spectra obtained.

126 Planetary surfaces as well as meteorites do not behave as Lambertian surfaces, meaning that
127 reflectance is strongly influenced by the illumination and observational geometries (Hapke, 1993; Beck
128 et al., 2012). This is expressed through anisotropy in the scattered light, depending on the optical
129 properties of the surface, its roughness, and the size, shape and porosity of the grains. Hence, variations
130 of the absolute measured reflectance and band depths can be observed for the same sample at different
131 observational geometries, while these parameters are commonly used in spectra description to
132 characterize a sample. The SuperCam instrument will achieve VNIR spectral measurements at various
133 observational geometries due to variations in solar incidence, observational geometry and local
134 topography, hence greatly restricting sample-to-sample comparison. To characterize the influence of
135 the observational geometry on the light scattered by one typical Martian meteorite, we additionally
136 measured the bidirectional reflectance spectra of the NWA 4766 sample at different observation angles,
137 both on a particulate and on a non-crushed rock sample.

138

139 **2 The Martian meteorites**

140

141 **2.1 Nomenclature**

142

143 Except for the breccia "Black Beauty" (NWA 7034 and pairings), Martian meteorites are usually
144 mafic to ultramafic magmatic rocks frequently showing cumulate textures. They were initially classified
145 into three main families based on the first observed falls Shergotty, Nakhla and Chassigny: the
146 shergottites, the nakhlites and the chassignites (SNCs), and later regrouped under the Martian

¹ Allan Hills

² Elephant Moraine

³ Miller Range

⁴ Northwest Africa

⁵ Dar al Gani

⁶ Sayh al Uhaymir

⁷ Caleta el Cobre

⁸ The sample selection reflects the current collection of Martian meteorites at the IPAG (Grenoble, France) and the ENS (Lyon, France) laboratories, with additional samples lent by the University of Brest and the IPGP (Paris, France).

147 denomination (Bogard and Johnson, 1983). The shergottites, the largest family in number of specimens,
148 mainly contain pyroxene (pigeonite and augite), plagioclase and some of them, olivine. Abundant
149 textural, mineralogical and compositional variability is observed within this family. In this study, the
150 following classification is adopted: "basaltic", "olivine-phyric", "olivine/orthopyroxene-phyric" and
151 "poikilitic" shergottites. Their petrology can be summarized as follows:

- 152 - the basaltic shergottites are fine-grained with doleritic texture, mainly composed of augite
153 and pigeonite clinopyroxenes and maskelynite (shocked plagioclase), and are devoid of
154 olivine (McSween, 1994);
- 155 - the olivine-phyric shergottites (hereafter abbreviated to ol-phyric shergottites) exhibit
156 olivine megacrysts, orthopyroxene and chromite, enclosed in a fine-grained groundmass of
157 pigeonite and maskelynite evidencing effusive texture (Goodrich, 2002);
- 158 - the olivine/orthopyroxene-phyric shergottites (hereafter abbreviated to ol/opx-phyric
159 shergottites) are similar to olivine-phyric shergottites, with additional orthopyroxene
160 megacrysts (Irving et al., 2004);
- 161 - the poikilitic shergottites were initially referred to as "lherzolitic shergottites" given their
162 high olivine content (McSween, 1994). They exhibit coarse-grained olivine enclosed in
163 orthopyroxene phenocrysts (i.e., a poikilitic texture), with interstitial groundmass filled by
164 pigeonite and augite clinopyroxenes, and to a lesser extent than in basaltic shergottites,
165 maskelynite. However, as exposed by Walton et al. (2012), they differ from the strict
166 definition of terrestrial lherzolites in that they are not plutonic rocks, and some of them have
167 too little olivine and too much plagioclase. Consistently, the terminology poikilitic
168 shergottite (Walton et al., 2012) is preferred here.

169 NWA 7635 is officially classified as a shergottite by the Meteoritical Society; it however differs
170 from the four shergottite groups described above. It exhibits plagioclase phenocrysts shocked to
171 maskelynite, olivine and augite, but no pigeonite or phosphate phase was observed, contrary to the
172 other shergottites (Lapen et al., 2017). Another classification of shergottites is based on their content in
173 light Rare Earth Elements (REE): "enriched", "intermediate" and "depleted", relatively to chondrites (e.g.,
174 Barrat et al., 2002; Borg et al., 2003; Basu Sarbadhikari et al., 2009). The second most abundant SNC
175 family, the nakhlites, consist of clinopyroxene cumulates. They are augite- and to a lesser extent olivine-
176 rich (for a review, see Treiman, 2005). Only three occurrences of chassignites, the dunite type of SNC,
177 were reported to date. In addition to the SNCs, three other meteorites were classified as Martian: a
178 polymict breccia (the biggest specimen being NWA 7034, Agee et al., 2013; Humayun et al., 2013;
179 Wittmann et al., 2015), the orthopyroxenite ALH 84001 (Mittlefehldt, 1994) and the augite-rich basalt
180 NWA 8159 (where pigeonite is absent unlike in shergottites, Ruzicka et al., 2015). The polymict breccia
181 NWA 7034 and pairings is a unique type of meteorite consisting of various lithic (norite, monzonite,
182 gabbro), mineral (feldspar, pyroxene, iron-oxides, apatite) and melt (rock and spherule) clasts, enclosed
183 in a fine-grained matrix enriched in magnetite and maghemite oxides (Agee et al., 2013; Humayun et
184 al., 2013).

185 Over the last decades, controversy has arisen on whether the shergottites are young (<1 Ga) or
186 old (>4 Ga) Martian rocks. On the one hand, analyses of individual minerals with the ^{87}Rb – ^{87}Sr ,
187 ^{147}Sm – ^{143}Nd , ^{176}Lu – ^{176}Hf and U–Pb chronology systems yield fairly young radiometric ages for the
188 shergottites (175–475 Ma; for a review, see Nyquist et al., 2001), interpreted as a crystallization during
189 the Amazonian era on Mars. On the other hand, bulk-rock analyses on the ^{207}Pb – ^{206}Pb and ^{87}Rb – ^{87}Sr
190 systems return contrasting radiometric ages at ~4.1 Ga (Bouvier et al., 2008). The shergottites' cosmic
191 ray exposure age points to ejections from Mars about ~0.5 Ma to ~20 Ma ago (Bogard et al., 1984;
192 Shukolyukov et al., 2002). Unlike the shergottites, there is little debate on the age of nakhlites and
193 chassignites, which share similar ejection and crystallization ages (~11 Ma and ~1.3 Ga respectively),
194 pointing to a single ejection event and a common origin on Mars (Bogard et al., 1984; for a review on
195 formation ages, see Borg and Drake, 2005). The oldest specimen is the orthopyroxenite ALH 84001,
196 whose ~4.5 Ga age yields a formation during the very early period of Mars (Nyquist et al., 1995).
197 Additionally, some zircons in the polymict breccia NWA 7533 have ages of ~4.4 Ga (Humayun et al.,
198 2013).

199

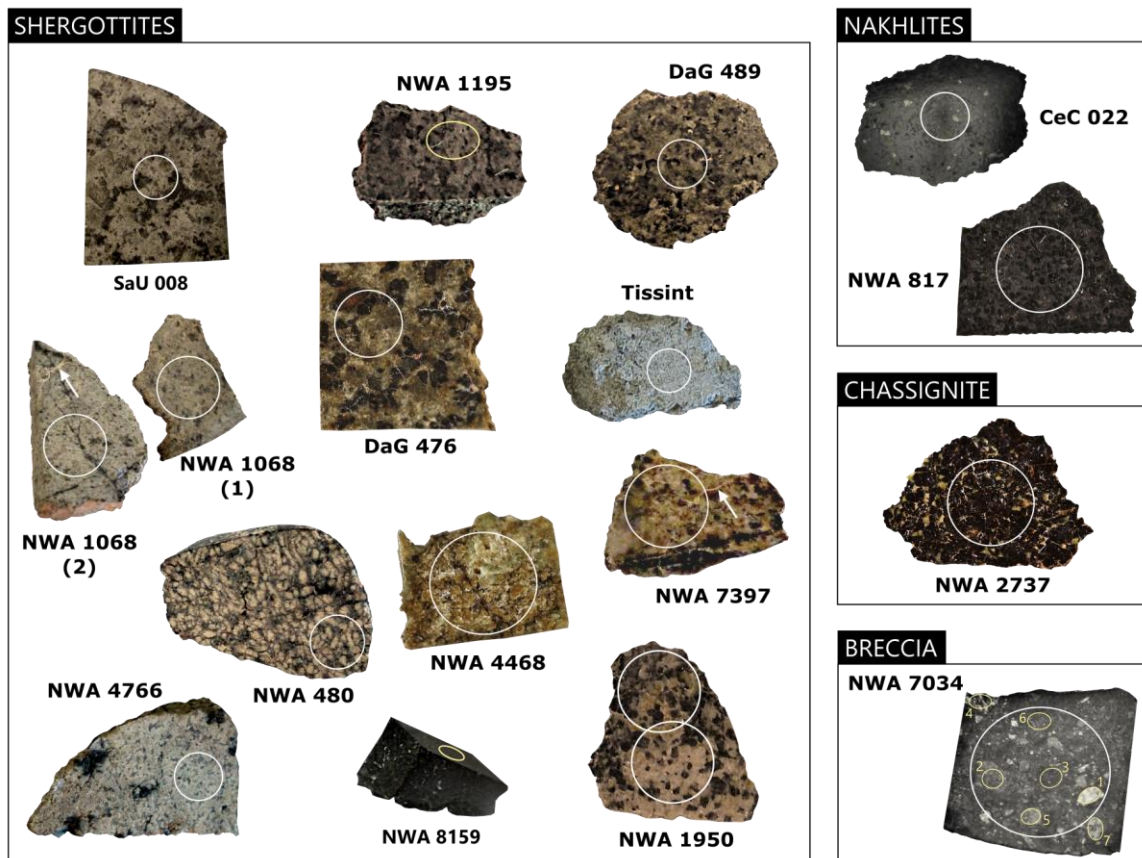
200 **2.2 Alteration minerals in Martian meteorites**

201

202 On Mars, orbital data as well as in situ measurements have shown the presence of various
203 alteration minerals: Fe/Mg and Al-smectites, kaolins, micas, serpentines, chlorites, prehnites, sulfates,
204 carbonates, zeolites, oxides and hydrated silica (Christensen et al., 2000; Bibring et al., 2005; Gendrin et
205 al., 2005; Ehlmann et al., 2008; Ehlmann et al., 2009; Carter et al., 2013; Vaniman et al., 2014). Martian
206 meteorites are predominantly composed of pyroxene, olivine and plagioclase (mainly in the form of
207 maskelynite), with minor accessory phases such as phosphates (apatite, merrillite). These primary
208 minerals are sometimes associated with alteration phases, of preterrestrial origin (formed on Mars or
209 during transport to the Earth) or produced during terrestrial residence. The latter occurs especially in
210 hot desert find meteorites, which were largely the focus of this study (e.g., Jull et al., 1990; Scherer et al.,
211 1992). Hence, it is likely that terrestrial weathering might play a significant role in forming any secondary
212 phase detected in the VNIR spectra presented in this study. The meteorites found in hot deserts are
213 usually more fractured than cold desert finds and can exhibit partial dissolution of primary minerals and
214 formation of secondary phases such as clay minerals, Fe-oxides/(oxy)hydroxides, carbonates, sulfates
215 and silica (Scherer et al., 1992; Lee and Bland, 2004).

216 In nakhlites, secondary minerals were reported in association with olivine and in the mesostasis.
217 These assemblages consist of Fe/Mg/Al-silicate amorphous phases, poorly-crystalline phyllosilicate
218 mixtures (likely containing smectite and serpentine), various carbonates, sulfates, salts, laihunite,
219 ferrihydrite and iron oxides (Gooding et al., 1991; McCubbin et al. 2009; Noguchi et al., 2009; Changela
220 and Bridges, 2010). Ca and Mg-carbonates as well as Ca-sulfates are also present in chassignites and
221 shergottites, but phyllosilicates rarely occur (Gooding et al., 1988; Gooding et al., 1991; Wentworth and
222 Gooding, 1994; Wentworth et al., 2005). In ALH 84001, corroded pyroxenes are associated with Ca, Fe
223 and Mg-carbonates in concentric disks, referred to as "rosettes" (Mittlefehldt, 1994). Except from ALH

224 84001, most of the carbonate assemblages in Martian meteorites occur in veins and are mostly terrestrial
 225 alteration products (Lee and Bland, 2004; Gillet et al., 2005; Treiman et al., 2007; Howart et al., 2014,
 226 Shearer et al., 2015). For example, Figure 1 shows a light, likely carbonate-filled veins spreading through
 227 the slab of shergottite NWA 7397 and NWA 1068 (2). Various oxides are reported in all Martian
 228 meteorites families (such as ilmenite, ulvöspinel, chromite and magnetite; for a review, see Papike et al.,
 229 2009). In particular, the polymict breccia NWA 7034 and pairs as well as the augite-rich basalt NWA 8159
 230 exhibit elevated content in iron oxides magnetite and maghemite (respectively ~15 wt% and ~6 wt%,
 231 Gattacceca et al., 2014; Herd et al., 2017). Similarly, the unusual dark color of the olivine grains in NWA
 232 2737 is attributed to the presence of nanophase kamacite alloy particles (Pieters et al., 2008).



233
 234 **Figure 1.** Martian meteorites non crushed rock suite and location of spectral measurements. The scale is given by
 235 the size of the measurement spot: white spots are ~5.2 mm large (regular measurement) and yellow spots are ~1.3
 236 mm by ~1.7 mm. Two spectra were obtained for NWA 1950, which exhibit a poikilitic (light region) and a non-
 237 poikilitic area (dark region; Gillet et al., 2005). Arrows indicate the carbonate-filled veins described in the text.

238
 239 **2.3 Absorption bands of the minerals of interest (in the 0.4-3 μm region)**

240
 241 *2.3.1 Absorption bands of primary minerals in Martian meteorites*

242 Martian meteorites are mostly composed of the mafic minerals olivine and pyroxene, as well as
 243 some shocked plagioclase in maskelynite form. These minerals present electronic absorptions in the

244 0.4–3 μm range caused by the presence of transition metals, the most intense absorptions being related
245 to iron, especially when in ferrous state (Fe^{2+}).

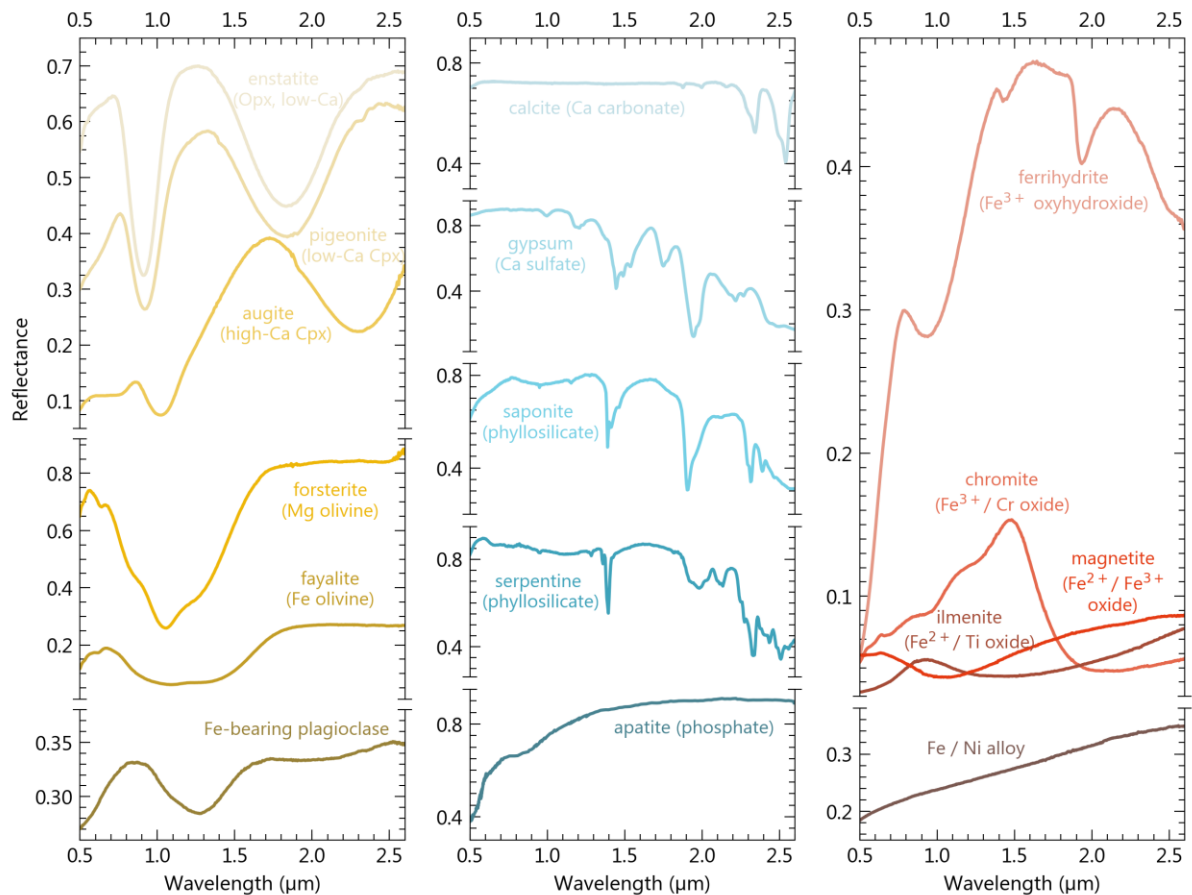
246 Pyroxenes ($(\text{Ca,Mg,Fe})_2\text{Si}_2\text{O}_6$) have reflectance spectra that exhibit two main absorption bands:
247 around $\sim 1 \mu\text{m}$ and $\sim 2 \mu\text{m}$, hereafter referred to as bands I and II (Adams, 1974; Fig. 2). The exact center
248 positions of these absorptions are greatly modulated by the substitution of cations in the crystal field,
249 here Fe, Mg and Ca (Adams, 1974; Cloutis and Gaffey, 1991; Klima et al., 2011). In clinopyroxenes (Cpx)
250 spectra, which show a wide variability in Ca content, band I and band II shift to longer wavelengths with
251 increasing Ca, with the shift of band I being subtler than that of band II (Adams et al., 1974; Fig. 2).
252 Among the major types of Cpx present in Martian meteorites, pigeonite is Ca-poor and has band I and
253 II usually positioned before 1 and 2 μm respectively, while augite is Ca-rich and has band I and II usually
254 positioned after 1 μm and 2.2 μm respectively (Fig. 2). Orthopyroxenes (Opx) are depleted in Ca: their
255 band I and II are usually positioned before 1 and 2 μm . Within a pyroxene family with fixed Ca content
256 such as the orthopyroxenes, the band center shift is mainly modulated by the Fe content, with
257 absorptions occurring at longer wavelength with increasing Fe (Cloutis and Gaffey, 1991).

258 The main absorption of olivine ($(\text{Mg,Fe})_2\text{SiO}_4$) is located in the $\sim 0.7\text{-}1.8 \mu\text{m}$ range and centered
259 around $\sim 1 \mu\text{m}$ (Fig. 2). The shape of the absorption is modulated by the Fe over Mg content: fayalite,
260 the Fe-endmember of olivine exhibits a wider and more flattened band than forsterite, the Mg-
261 endmember (King and Ridley, 1987; Fig. 2).

262 Visible and near-infrared spectra of plagioclase feldspar ($(\text{Na,Ca})(\text{Si,Al})_4\text{O}_8$), when Fe-bearing,
263 are characterized by a weak absorption in the $0.8\text{-}1.7 \mu\text{m}$ range and centered around $\sim 1.1\text{-}1.3 \mu\text{m}$,
264 deepening with increasing Fe (Adams and Goullaud, 1978; Fig. 2). Of relevance for maskelynite
265 identification in reflectance data, shocked plagioclase experiments show a drop of the absolute
266 reflectance and a decrease in the absorption band strength with increasing shock pressures (Johnson
267 and Hörz, 2003). Mixed with pyroxene or olivine, plagioclase is usually detected only if present in high
268 abundance and rich in iron (the limit of detection depends on the grain size; Crown and Pieters, 1987).

269 The iron and nickel alloys detected in Martian meteorites absorb efficiently at VNIR wavelengths
270 and usually show a red slope in this range (Fig. 2). Finally, phosphates can exhibit various absorption
271 bands related to Fe electronic processes if iron-bearing, and vibrations of the PO_4^{3-} , H_2O and OH groups
272 if hydrated or hydroxylated. Merrillite and apatite are depleted in iron; hence, if neither hydrated nor
273 hydroxylated, their spectra are relatively featureless, the PO_4^{3-} bands being usually very subtle (for a
274 review, see Bishop et al., 2019; Fig. 2).

275



276

277 **Figure 2.** Laboratory reflectance spectra of various minerals reported in Martian meteorites and absorbing in the
 278 VNIR range, mainly taken from the PDS Geoscience Spectral Library. Spectra IDs are as follows: enstatite C2PE30,
 279 augite C1SC35, pigeonite C1PP42, forsterite C3PO51, fayalite C3PO59, plagioclase NCLS04, calcite CBCB12, gypsum
 280 F1CC16B, saponite LASA51, serpentine LASR10, ferrihydrite C1JB45, chromite LACR12, magnetite LAMG07, ilmenite
 281 LAPI06A and Fe/Ni alloy C1SC12. Apatite spectrum is taken from Lane et al. (2007). The absolute reflectance values
 282 are conserved in the plots; however, we encourage the reader to keep in mind that these values are dependent from
 283 the sample grain size adopted in the spectral measurement (e.g., Salisbury and Wald, 1992; Mustard and Hays,
 284 1997).

285

286 2.3.2 Absorption bands of alteration minerals in Martian meteorites

287 Infrared spectrometry is a powerful tool to assess the presence of alteration minerals. In
 288 particular, combination of bending and stretching of groups such as H_2O , OH^- , metal-OH, CO_3^{2-} and
 289 SO_4^{2-} in minerals are responsible for the majority of the vibrational absorptions observed in the near-
 290 infrared for the alteration minerals reported in Martian meteorites. The water molecule and the hydroxyl
 291 ion produce an intense absorption band centered at $3 \mu\text{m}$ as well as two narrow bands at $1.4 \mu\text{m}$ and
 292 $1.9 \mu\text{m}$ (Hunt, 1977; Fig. 2). These two absorptions usually require the presence of water at a structural
 293 level in minerals to appear, while the $3 \mu\text{m}$ band is commonly observed in laboratory spectral
 294 measurements and does not require large amounts of water to appear (e.g., if adsorbed water is present
 295 on the sample). Also of relevance for the hydrous minerals detection, hydroxyl bounded to metal cations

296 (such as $\text{Fe}^{2+/3+}$, Mg^{2+} , Al^{3+} , Si^{4+} , etc.), creates additional narrow absorptions in the near-infrared (Hunt,
297 1977; Fig. 2). The phyllosilicate assemblages reported in nakhlites (i.e., smectite and serpentine) present
298 these kinds of absorptions (Fig. 2). Carbonates have specific absorptions related to the CO_3^{2-} ion, which
299 appear in the 0.4-3 μm region at $\sim 2.3 \mu\text{m}$ and $\sim 2.5 \mu\text{m}$ (Hunt and Salisbury, 1971; Fig. 2). Sulfate minerals
300 have additional specific absorptions related to the SO_4^{2-} ion, mostly between $\sim 2.1 \mu\text{m}$ and $2.7 \mu\text{m}$
301 (Cloutis et al., 2006; Fig. 2). Finally, the iron-oxides occurring in meteorites present various broad
302 absorptions in the VNIR resulting from the electronic processes related to iron (Sherman et al., 1982;
303 Fig. 2). These oxides are very efficient at absorbing the VNIR light, with absolute reflectance commonly
304 below 15% (Fig. 2). As a matter of fact, the unique darkness of the polymict breccia NWA 7533 has been
305 attributed to its elevated magnetite and maghemite contents (Beck et al., 2015).

306

307 **3 Samples and methods**

308

309 **3.1 Meteorite suite**

310

311 We gathered a sample suite representative of the Martian meteorite's diversity described in section
312 2.1 (Fig. 1 and 3). This suite comprises:

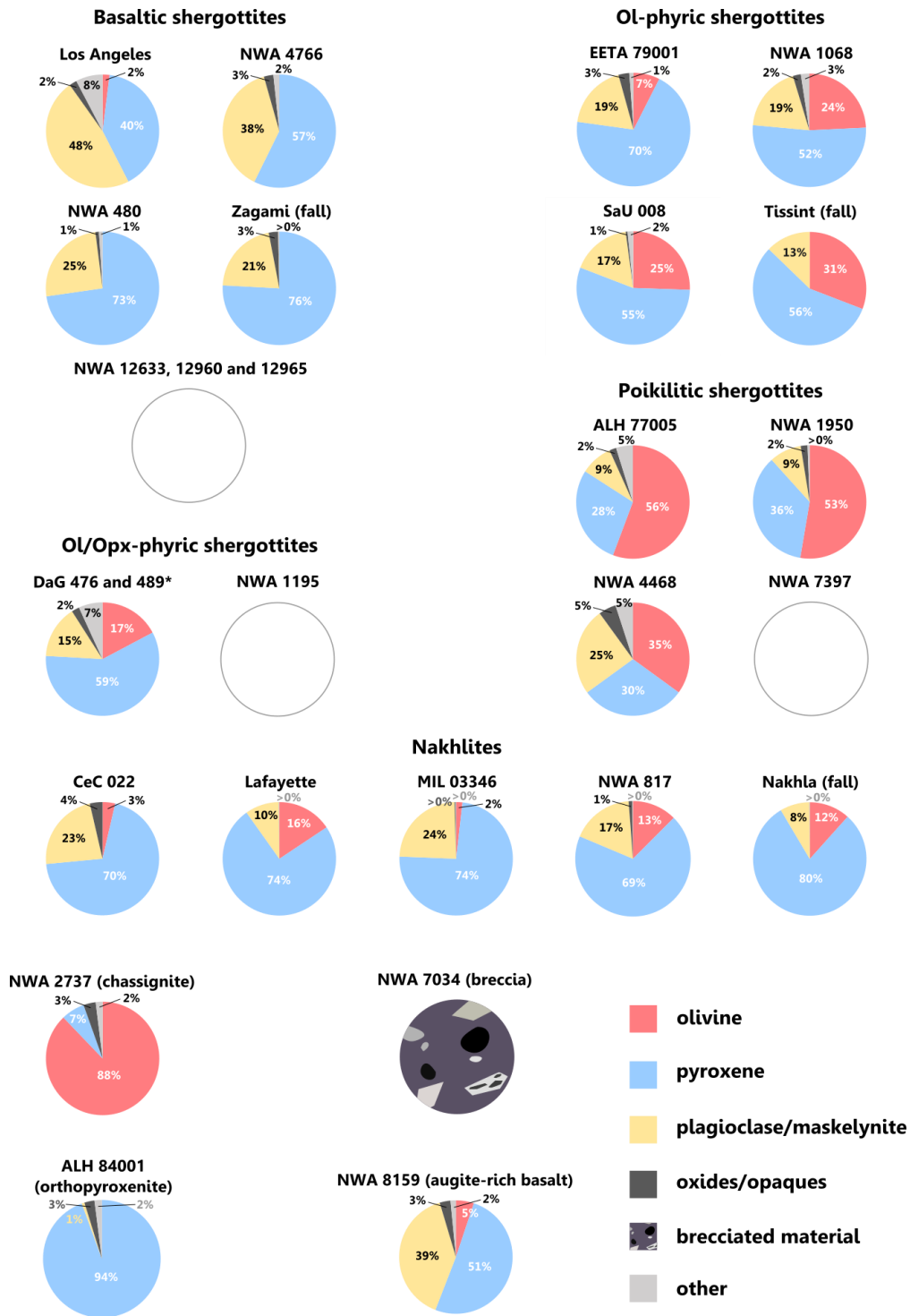
- 313 - The orthopyroxenite ALH 84001;
- 314 - The polymict breccia NWA 7034;
- 315 - The augite-rich basalt NWA 8159;
- 316 - One out of the 3 chassignites NWA 2737;
- 317 - Five out of the 13 nakhlite pairings⁹ (see Fig. 3 for sample names);
- 318 - 18 shergottites out of the 204 individual specimens (with likely pairings) approved by the
319 Meteoritical Society, including all the main classes (see Fig. 3 for sample names):
 - 320 o Seven basaltic shergottites;
 - 321 o Three olivine/orthopyroxene-phyric shergottites (including at least a pairing);
 - 322 o Four olivine-phyric shergottites;
 - 323 o Four poikilitic shergottites.

324 The locations of the spectral measurements on the rock samples are shown in Figure 1.

325

326 Figure 3 shows the modal mineralogy of these meteorites (values retrieved from the literature).
327 Consistently with the Martian meteorite global diversity and except some effusive ol/opx- and ol-phyric
328 shergottites, most of our samples are consistent with cumulates hypovolcanic rocks (i.e., excavated from
329 depth), as indicated by the predominance of pyroxene and olivine. Here, the only meteorite that exhibits
330 a modal mineralogy consistent with a surface basalt resulting from effusive processes from a non-
331 fractioned primary melt is Los Angeles.

⁹ As of 2020



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Fig 3. Classification of the Martian meteorites analyzed and modal mineralogy retrieved from the literature (averaged from Stolper & McSween 1979; Ma et al., 1981; Mason 1981; Treiman et al., 1994; Gleason et al., 1997; Lentz et al., 1999; Folco et al., 2000; Rubin et al., 2000; Zipfel et al., 2000; Mikouchi 2001; Wadhwa et al., 2001; Barrat et al., 2002; Gillet et al., 2002; Mikouchi & Miyamoto 2002; Sautter et al., 2002; Xirouchakis et al., 2002; Bartoschewitz et al., 2003; Russell et al., 2004; Anand et al., 2005; Gillet et al., 2005; McKay & Schwandt 2005; Mikouchi et al., 2005; Rutherford et al., 2005; Stopar et al., 2005; Beck et al., 2006; Day et al., 2006; Wittke et al., 2006; Imae & Ikeda 2007; Irving et al., 2007; Treiman et al., 2007; Walton and herd 2007; Lapen et al., 2010; Hsu et al., 2012; Ruzicka et al., 2015; Herd et al., 2017; Krämer Ruggiu et al., 2019). Empty circles correspond to lack of values.

342 * DaG 476 and DaG 489 are likely paired together (Grossman, 1999).

343

344 Some of the cut section samples were covered by resin in the past, which was removed by thin polishing
345 of the sections by previous owners. As the resin percolated through the grains of the samples, a removal
346 of this leftover resin without a destructive method could not be achieved. These residues can appear in
347 our spectra as contaminants, but their spectral signatures do not affect the major mineral bands and are
348 not an issue for identifying the main mineralogy in the point spectra and hyperspectral cubes. However,
349 some types of resin can exhibit absorption bands similar to those of secondary minerals, at 1.9 μm or
350 2.3 μm and 2.5 μm (e.g., Li et al., 2017). This issue was considered when interpreting the various point
351 spectra and hyperspectral cubes of cut sections mounted in resin.

352

353 **3.2 Point spectra acquisition**

354

355 The point reflectance spectra of the meteorites were acquired using the Spectrophotometer
356 with cHanging Angles for the Detection Of Weak Signals (SHADOWS, Potin et al., 2018) designed at the
357 Institut de Planétologie et d'Astrophysique de Grenoble (IPAG). SHADOWS is a spectro-goniometer
358 capable of acquiring individual spectra of dark samples ($>0.005\%$ in reflectance) at various geometries
359 of illumination and observation angles (Potin et al., 2018). The light source is located on the first arm of
360 the goniometer and consists of a monochromatic beam transported through eight optical fibers and
361 focused on the sample. The reflected light is collected by two detectors, one covering the VNIR range
362 from 0.185 μm to 1.2 μm , the other covering the infrared range from 0.8 μm to 5.2 μm , and located on
363 a second arm (Potin et al., 2018). The Martian meteorites spectra were measured at a spectral sampling
364 of 20 nm and a variable spectral resolution (on the order of a few tens of nanometers, Potin et al., 2018)
365 mainly between 0.36 μm and 3 μm , as this range contains the majority of absorption bands of the
366 minerals of interest (both primary and secondary phases; e.g., Singer et al., 1981; Ody et al., 2012; Carter
367 et al., 2013; Fig. 2).

368

369 *3.2.1 Primary spectral measurements*

370 We performed measurements on all the samples at a phase angle $\phi = 30^\circ$, corresponding to an
371 incidence angle θ_i of 0° (illumination at nadir) and an emergence (observation) angle θ_e of 30° . The
372 incidence and emergence angles θ_i and θ_e are here defined as the angle between the normal to the
373 surface and the incident beam, and between the normal to the surface and the observed direction,
374 respectively. The phase angle ϕ corresponds to the angle between these two directions. At the geometry
375 of measurement, the resulting illumination spot size is ~ 5.2 mm wide (Potin et al., 2018; Fig. 1). For
376 smaller samples, a pinhole of 500 μm is placed to select the incident beam of only one optical fiber
377 while masking the seven others, resulting in a ~ 1.3 mm by ~ 1.7 mm illumination spot and lower signal-
378 to-noise (SNR) ratio (Potin et al., 2018; Fig. 1). For each day of measurement, spectra of reference targets
379 (SpectralonTM and InfragoldTM) are acquired and used to calibrate the data. Additional software
380 correction of the data also includes adjustments related to the photometric angle used in the

381 measurements (e.g., modification of the illumination spot shape, non-Lambertian behavior of the
 382 spectralon, see Potin et al., 2018).

383

384 *3.2.2 Spectral measurements with varying observation geometry*

385 We characterized the anisotropy of the light scattered by NWA 4766, both on powder and cut
 386 section, by acquiring bidirectional spectral measurements between 0.36 μm and 3 μm with the point
 387 spectrometer SHADOWS, at various incidence, emergence and azimuth angles. This sample was selected
 388 because it is part of an abundant type of Martian meteorites, the basaltic shergottites (see section 2.1),
 389 and because it shows no obvious indication of terrestrial alteration (e.g., no vein crosscutting the melt
 390 veins and pockets, no absorption band related to the presence of secondary minerals in the sample).
 391 The powder was prepared by crushing a few milligrams of NWA 4766 in an agate mortar. Similar to the
 392 sample preparation detailed in Beck et al. (2012), the powder was not sieved, as a means to produce a
 393 particulate sample similar to a natural particulate medium, with various grain sizes (on the order of tens
 394 of microns maximum). To ensure accurate similitude of the measurements on the rock and the powder,
 395 homogeneous areas of the meteorite were considered for the spectra acquisition on the chip, and for
 396 sampling of the area to be crushed for powder preparation.

397 Measurements at positive azimuth angles (outside of the principal plane) were performed by
 398 rotating the observation arm in the horizontal plane. High phase angle measurements are limited by the
 399 extent of the measured surface and consequently the quantity of available meteorite material (i.e., high
 400 phase angle measurements require a wide surface as a result of the illumination spot elongation). No
 401 measurement was made at phase angles lower than 10° , as in this configuration, parts of the goniometer
 402 illumination arm are masking the illumination spot from the detector. The geometries of measurements
 403 explored are given in Table 1.

404

405 **Table 1.** List of observational geometries used for bidirectional spectral measurements on Martian meteorite NWA
 406 4766 (basaltic shergottite).

Sample	Incidence θ_i	Emergence θ_e	Azimuth a	Min. phase angle ϕ	Max. phase angle ϕ
NWA 4766 (powder)	$0^\circ, 20^\circ, 40^\circ, 60^\circ$	$-60^\circ, -50^\circ, -40^\circ, -30^\circ, -20^\circ, -10^\circ,$ $0^\circ, 10^\circ, 20^\circ, 30^\circ, 40^\circ, 50^\circ, 60^\circ$	0°	10°	120°
NWA 4766 (cut section)	$0^\circ, 40^\circ$	$-50^\circ, -40^\circ, -30^\circ, -20^\circ, -10^\circ,$ $0^\circ, 10^\circ, 20^\circ, 30^\circ, 40^\circ, 50^\circ$	$0^\circ, 44^\circ$	10°	90°

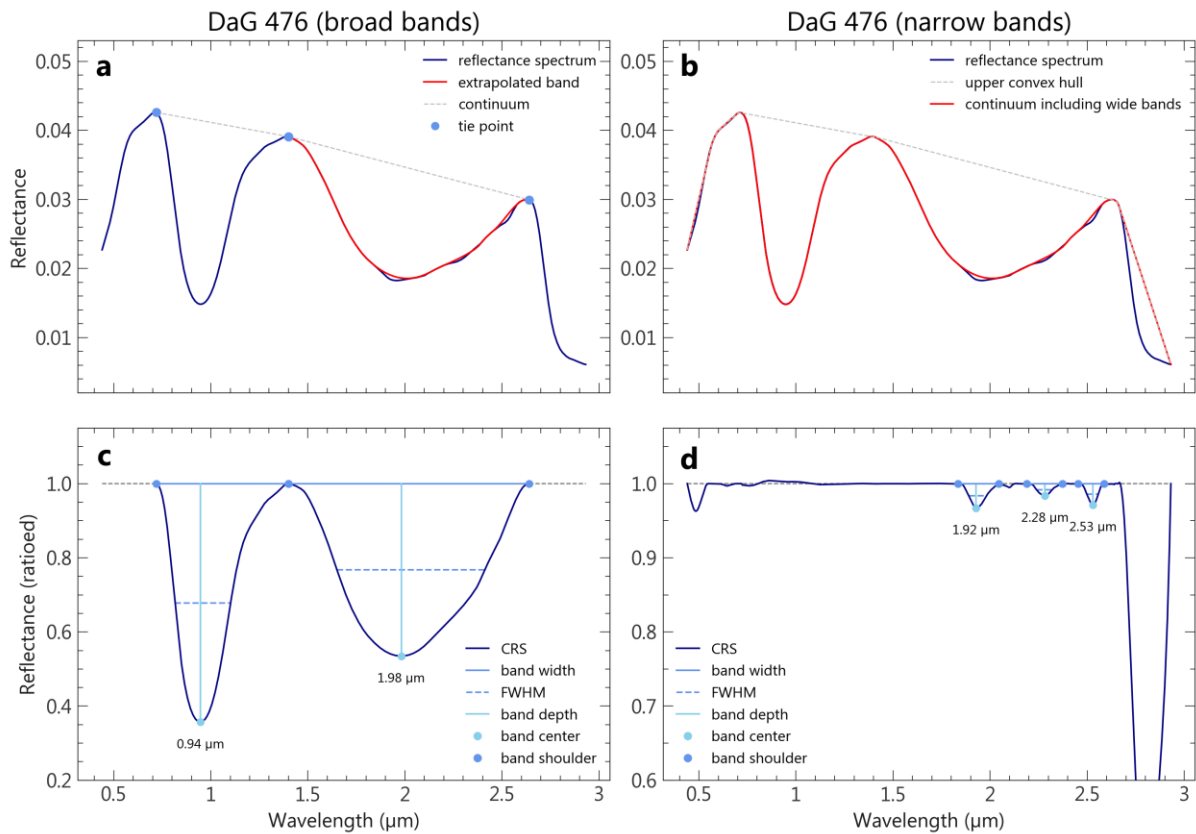
407

408 **3.3 Band parametrization**

409

410 To fully describe the absorption bands of each meteorite's point spectrum, we use the following
 411 parameters: band position center, band width, band Full Width at Half Maximum (FWHM) and band
 412 strength (or band depth, abbreviated BD). In order to retrieve these band parameters, we first perform
 413 continuum removal, using the following procedure:

- 414 - For spectra showing only the two main absorption bands of pyroxene at $\sim 1 \mu\text{m}$ and $\sim 2 \mu\text{m}$, a
415 spectral continuum is estimated for each of these two bands, similarly to the method used by
416 Horgan et al. (2014) and Martinot et al. (2018). For each band, a set of potential linear
417 continuums defined by a series of potential tie points are computed: the optimized continuum
418 is the one which maximizes the area of the absorption band (Fig. 4a). For the band centered at
419 $\sim 1 \mu\text{m}$, left shoulder tie points are set between $0.6 \mu\text{m}$ and $0.9 \mu\text{m}$ and right shoulder between
420 $1 \mu\text{m}$ and $1.5 \mu\text{m}$. For the band centered at $\sim 2 \mu\text{m}$, left shoulder tie points are set between 1.1
421 μm and $1.9 \mu\text{m}$ and right shoulder between $2.2 \mu\text{m}$ and $2.7 \mu\text{m}$. Visual examination of the best-
422 modelled continuum is performed, and the tie points values are adjusted if required. The
423 continuum-removed bands are then computed by dividing bands I and II by their respective
424 modelled linear continuum (Fig. 4c).
- 425 - For spectra with narrow absorption bands but no broad bands related to mafic minerals, an
426 upper convex hull is computed and divided from the original spectrum. If needed, this process
427 is repeated until all the absorption bands are correctly wrapped in the hull.
- 428 - In the case of spectra exhibiting narrow bands superimposed on the broad bands of mafic
429 minerals, a first continuum-removed spectrum (CRS) is obtained by maximizing the pyroxene
430 band area (first method described above). The two bands of pyroxene without the superimposed
431 bands are modelled by removing parts of the original spectra where narrow bands are observed,
432 and filling these parts using cubic interpolation (Fig. 4b). The resulting spectrum is used to
433 retrieve parameters of the bands corresponding to pyroxene. In a second step, the modelled
434 broad bands are divided from the original spectrum to retrieve the narrow superimposed
435 absorption bands (Fig. 4d).
- 436



437

438 **Figure 4.** Spectral analysis performed to retrieve band parameters, illustrated for Martian meteorite DaG 476. **(a)**
 439 The continuum line is estimated by maximizing each band area between variable tie points. When narrow
 440 absorption bands are superimposed to a broad band of pyroxene, the corresponding broad band is extrapolated
 441 above the narrow bands. **(b)** When narrow absorptions are present, an additional continuum which is composed of
 442 the broad bands and an upper convex hull outside of the broad bands is computed. By doing so, both bands
 443 superimposed to the broad bands and bands located outside of the broad bands can be correctly detected. **(c)**
 444 Parametrization of the wide absorptions related to pyroxene. The CRS is obtained by dividing the original spectrum
 445 of DaG 476 corrected from the presence of the narrow absorption bands by the continuum shown in (a). **(d)**
 446 Parametrization of the narrow bands related to secondary minerals. The slight convexity in the spectrum causing
 447 the detection of a band at 0.48 μm is not considered in this study, hence the band was manually ignored. The CRS
 448 is obtained by dividing the original spectrum of DaG 476 by the continuum including the wide absorptions of
 449 pyroxene shown in (b).

450

451 Band parameters can be derived from the CRS if the left and right shoulders of the absorptions are
 452 known. For the two main bands of pyroxene, they correspond to the tie points of the optimized linear
 453 continuum (Fig. 4a and 4b). Aside from this case, a band is defined if a local minimum is found on the
 454 continuum-removed spectrum; the band shoulders are defined as the local maxima closest to this point.
 455 To exclude artificial band detection caused by noise, a threshold on the minimum absorption strength
 456 is set (usually 0.5% below the continuum baseline and adjusted if needed). Finally, the absorption bands
 457 are parameterized as follows (Fig. 4c and d):

- 458 - The band position, or band center is expressed by the wavelength at which the minimum CRS
- 459 value occur, in the spectral range bounded by the band shoulders;
- 460 - The band depth is equal to 1 minus the CRS value at the band center;
- 461 - The width is defined as the right shoulder position minus the left shoulder position (in microns);
- 462 - The FWHM corresponds the width of the band at half of the band depth.

463

464 **3.4 Hyperspectral cubes acquisition**

465

466 In addition to point spectrometry, we analyzed some of the meteorite samples using imaging
467 spectrometry, where hyperspectral images of the samples, or cubes, are produced, with the third
468 dimension corresponding to the wavelength axis. We imaged DaG 476, DaG 489, NWA 480, NWA 1068,
469 NWA 1195, NWA 1950, NWA 2737, NWA 4468 and NWA 7034 using a HySpex SWIR-384 imaging
470 spectrometer. The unprocessed samples are placed on a translating table, allowing line-by-line scanning
471 of the whole spectrum, and illuminated by a 0.4–2.5 μm broadband halogen light source at an angle of
472 $\sim 30^\circ$ as they pass through the field of view of the detector. Hyperspectral images are acquired in 288
473 bands sampled every 5.45 nm and ranging from 0.93 to 2.50 μm , with a spatial resolution of ~ 50
474 $\mu\text{m}/\text{pixel}$. Additional cubes in the near-infrared were acquired on SaU 008 and Tissint at the University
475 of Bern with the imaging system of the SCITEAS facility (Pommerol et al., 2015). Here, a narrow bandpass
476 of light is selected by a monochromator coupled to a halogen source and illuminates the samples, while
477 the reflected photons are collected by a camera placed at nadir and covering the near-infrared range
478 between 0.82 and 2.5 μm . Unlike the HySpex spectrometer, a complete image of the whole sample is
479 produced for every time step, one wavelength at a time. The light's wavelength was changed between
480 each image using 6 nm steps, resulting in hyperspectral cubes similar to the HySpex products in terms
481 of spectral range and sampling. The spatial resolution (~ 0.5 mm/px) is lower than for the HySpex
482 measurements but remains sufficient to resolve the main phenocrysts of Tissint and SaU 008 meteorites.
483 Internal relative calibration for both experiments is achieved using a SpectralonTM reflectance target.

484 To guide the spectral analysis and interpretation of the cubes, spectral index maps are produced
485 based on the spectral features of the main absorbing minerals present in meteorites: olivine, low-calcium
486 pyroxene (LCP, e.g., orthopyroxene and pigeonite) and high-calcium pyroxene (HCP, e.g., augite). Olivine
487 is mapped using the olivine index described in Mandon et al. (2020): the depth of four absorption bands
488 at 1.08 μm , 1.26 μm , 1.37 μm and 1.47 μm are measured, as well as the convexity at 1.3 μm (Mg-olivine)
489 and 1.4 μm (Fe-olivine and coarse-grained Mg-olivine). If three out of the four absorption bands and
490 one out of the two convexities are positive, the value of the olivine parameter corresponds to the
491 average of the band depths. The two types of pyroxenes are mapped using an adaptation of the
492 "*LCPINDEX2*" and "*HCPINDEX2*" parameters from Viviano-Beck et al. (2014), which are weighted
493 averages of the depths of selected bands between 1.69 μm and 1.87 μm for the LCP index, and between
494 2.12 μm and 2.46 μm for the HCP index.

495

496 **4 Spectral features of Martian meteorites**

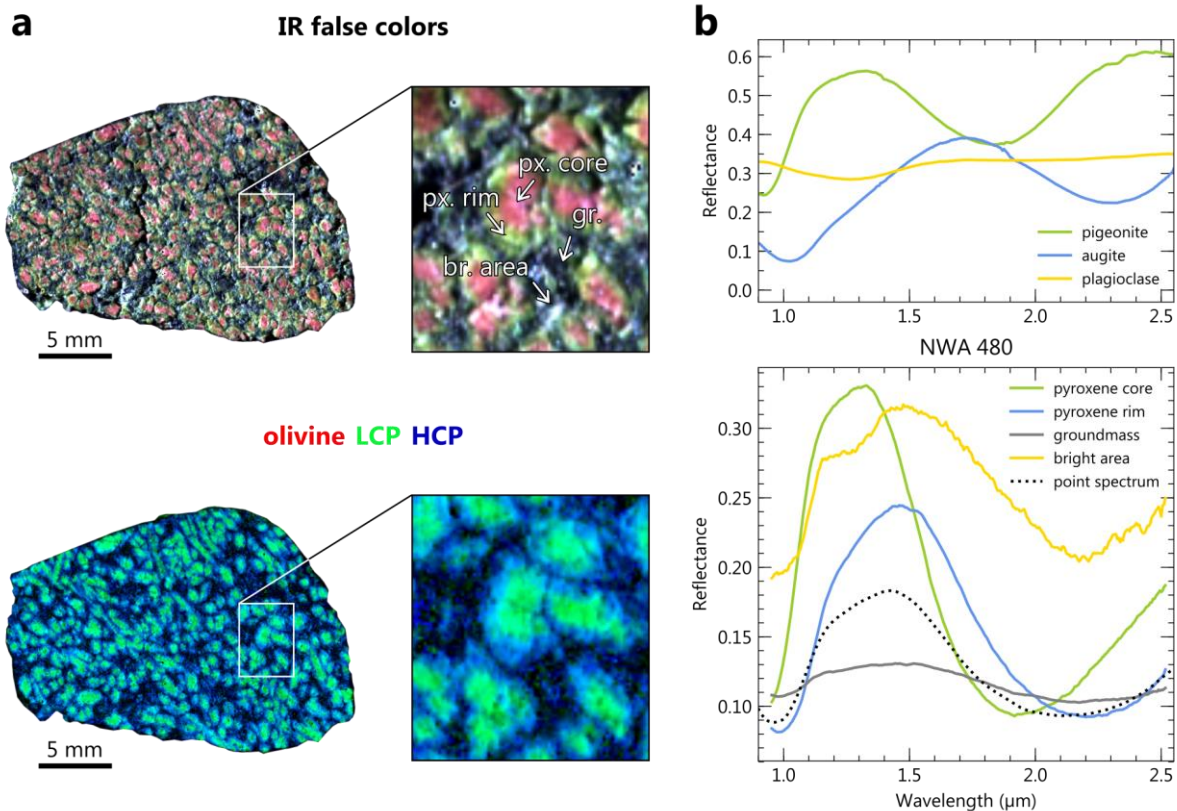
497

498 **4.1 Results from hyperspectral cubes**

499

500 False color composites in the near-infrared, parameters maps and spectra extracted from the
501 hyperspectral cubes are shown in Figures 5 to 9. Olivine, LCP and HCP are easily identifiable both in the
502 HySpex and SCITEAS infrared imaging products and they respectively appear blue, red/pink and
503 green/yellow in our false color RGB composite (R = 1.22 μm , G = 1.66 μm , B = 2.15 μm). Spectral
504 parameter maps are also successful at identifying these minerals, as confirmed by the various spectra
505 extracted from the cubes (Fig. 5 to 8). Augite and pigeonite individual crystals or zoning are resolved for
506 the basaltic and poikilitic shergottites, chassignite NWA 2737 and breccia NWA 7034 (Fig. 5 to 9). Even
507 at the high spatial resolution of HySpex products, the 1.3 μm absorption feature in Fe-bearing
508 plagioclase or maskelynite is not definitively detected, except for the basaltic shergottite NWA 480
509 where it might occur as bright patches in the groundmass, in association to HCP (Fig. 5b). The ol-phyric
510 and ol/opx-phyric shergottites exhibit dark regions of recrystallized impact melt pockets, characterized
511 by generally flat spectra and reflectance values lower than 2% in the VNIR (Fig. 6). The olivine crystals in
512 NWA 2737 that appear extremely dark in the visible (Fig. 1) also exhibit very low reflectance in the near-
513 infrared and a moderate red slope (Fig. 8). This is in agreement with previous studies proposing the
514 presence of Fe/Ni alloy in olivine crystals to be responsible for their unusual dark color (e.g., Van de
515 Moortèle et al., 2007; Pieters et al., 2008).

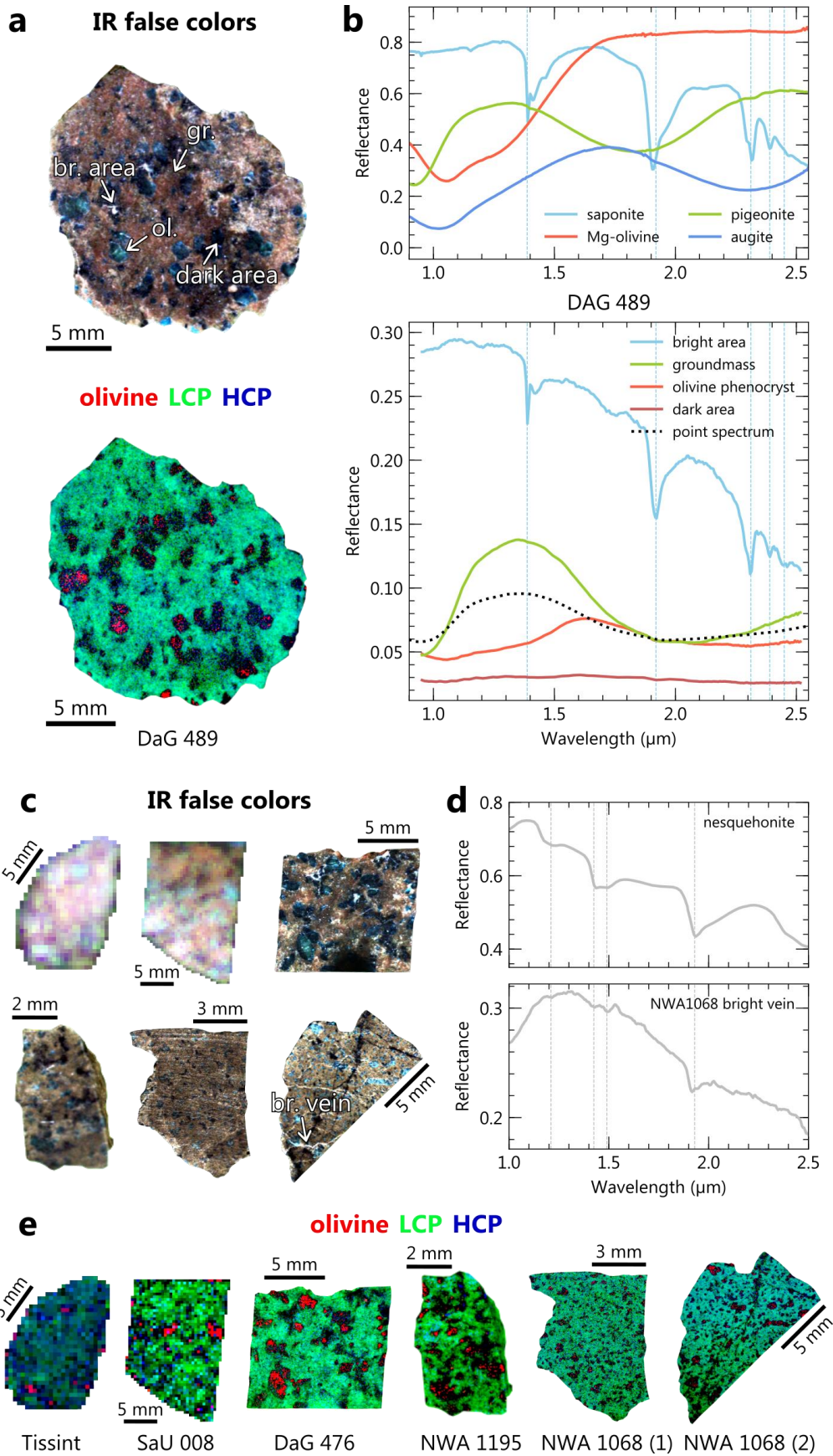
516



517

518 **Figure 5.** Hyperspectral cube of the basaltic shergottite NWA 480. **(a)** False RGB color composite in the near-
 519 infrared ($R = 1.22 \mu\text{m}$, $G = 1.66 \mu\text{m}$, $B = 2.15 \mu\text{m}$). In this composition, mineral zoning from LCP (pink) to HCP (green)
 520 is clearly visible. This is confirmed through examination of the RGB spectral parameters map below, where $R =$
 521 olivine index, $G =$ LCP index and $B =$ HCP index, and examination of the individual spectra. **(b)** Spectra extracted
 522 from the hyperspectral cube of NWA 480 (solid lines, location of spectral extraction shown with arrows on panel
 523 (a)) and point spectra obtained with the point spectrometer on a large part of the sample surface (dotted lines, see
 524 Fig. 1 for measurement location), compared with laboratory reflectance spectra of known minerals taken from the
 525 PDS Geoscience Spectral Library. Laboratory spectra IDs are as follows: pigeonite C1PP42, augite C1SC35 and
 526 plagioclase NCLS04. The pyroxene core and pyroxene rim spectra are consistent with LCP (represented by pigeonite)
 527 and HCP (represented by augite), respectively. The groundmass, though less reflective, has a spectrum with band
 528 centers roughly intermediate between those of low and high-calcium pyroxenes. The bright area spectrum
 529 resembles that of the groundmass, with increased reflectance level and an additional absorption at $\sim 1.3 \mu\text{m}$
 530 consistent with a mixture with plagioclase.

531



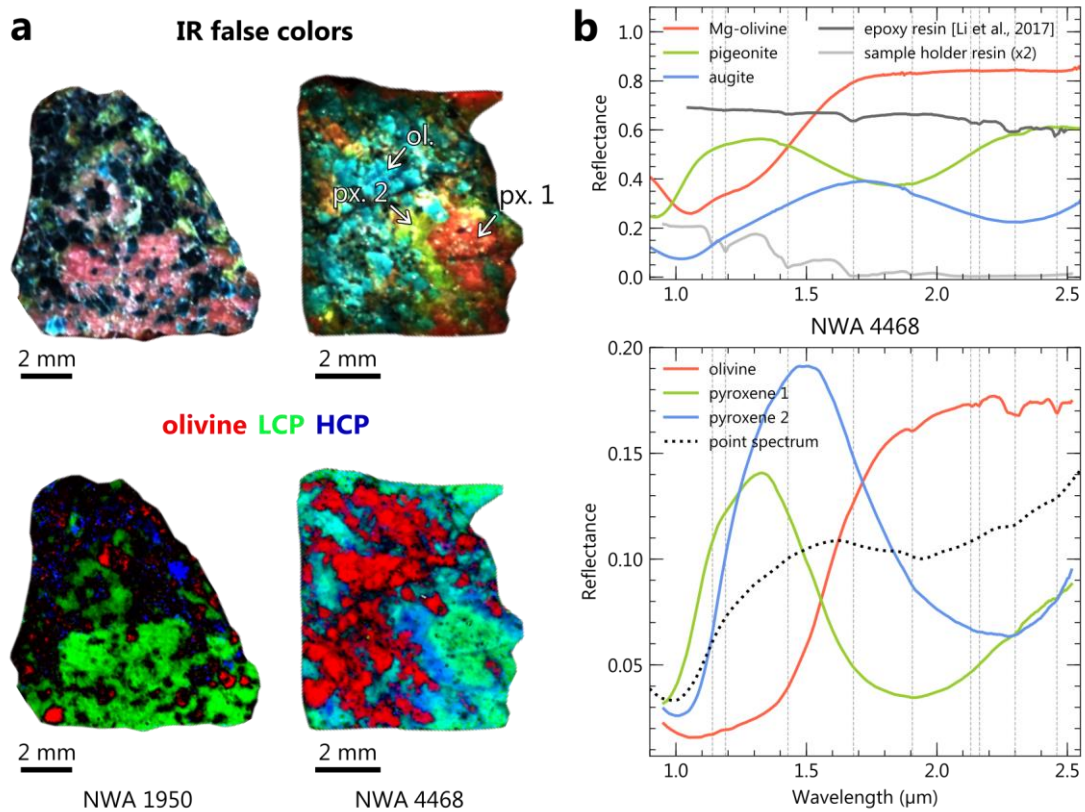
532

533 **Figure 6.** Hyperspectral cube of ol/opx-phyric and ol-phyric shergottites. **(a)** False RGB color composite of DaG 489

534 in the near-infrared ($R = 1.22 \mu\text{m}$, $G = 1.66 \mu\text{m}$, $B = 2.15 \mu\text{m}$) and associated RGB spectral parameters map below

535 (R = olivine index, G = LCP index, B = HCP index). In the IR false color composite chosen, olivine, LCP and HCP
 536 appear blue, red/pink and green/yellow respectively. **(b)** Spectra extracted from the hyperspectral cube of DaG 489
 537 (solid lines, location of spectral extraction shown with arrows on panel (a)) and point spectra obtained with the
 538 point spectrometer on a large part of the sample surface (dotted lines, see Fig. 1 for measurement location),
 539 compared with laboratory reflectance spectra of known minerals taken from the PDS Geoscience Spectral Library.
 540 Laboratory spectra IDs are as follows: saponite LASA51, forsterite C3PO51, pigeonite C1PP42 and augite C1SC35.
 541 DaG 489 olivine phenocrysts have spectra typical of Mg-olivine. The groundmass spectrum exhibits absorption
 542 centers intermediate between low and high-calcium pyroxenes, closer to the LCP endmember. The bright patches
 543 filling the groundmass and phenocryst voids have spectra consistent with a Mg-smectite (best matched by saponite
 544 smectite). **(c)** False RGB color composite of other ol/opx-phyric and Ol-phyric shergottites in the near-infrared (R =
 545 1.22 μm , G = 1.66 μm , B = 2.15 μm). **(d)** Spectrum extracted from a bright vein of NWA 1068 (2), compared with a
 546 spectrum of nesquehonite (hydrated Mg-carbonate) measured by Harner and Gilmore (2015). **(e)** RGB spectral
 547 parameters map (R = olivine index, G = LCP index, B = HCP index) associated with the meteorites in (c).

548

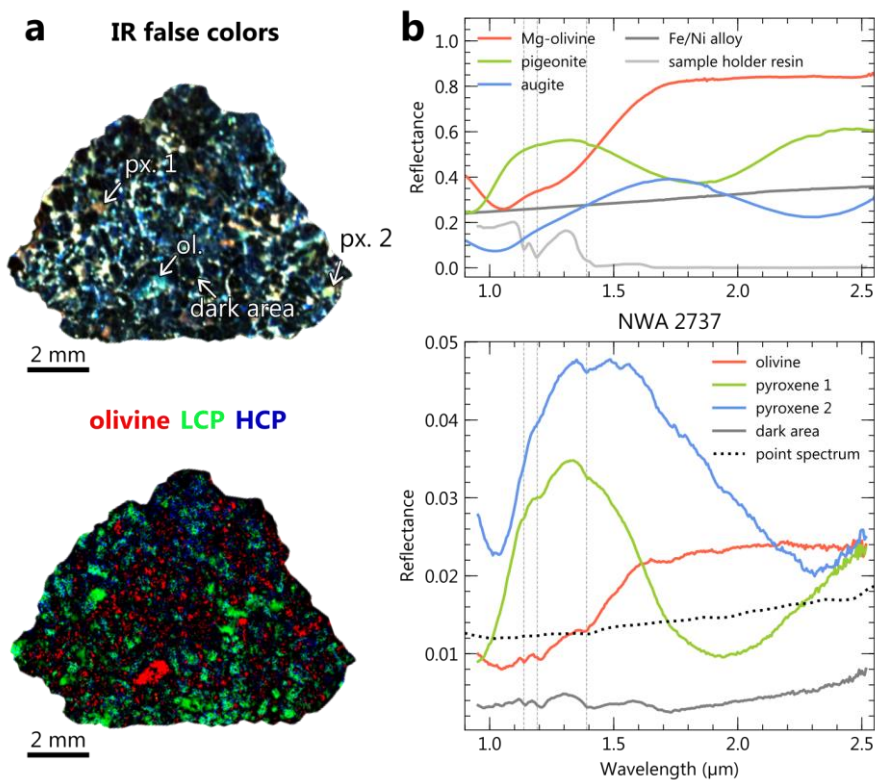


549

550 **Figure 7.** Hyperspectral cube of poikilitic shergottites. **(a)** False RGB color composite in the near-infrared (R = 1.22
 551 μm , G = 1.66 μm , B = 2.15 μm) and associated RGB spectral parameters map below (R = olivine index, G = LCP
 552 index, B = HCP index). In the IR false color composite chosen, olivine, LCP and HCP appear blue, red/pink and
 553 green/yellow respectively. **(b)** Spectra extracted from the hyperspectral cubes of NWA 1950 and NWA 4468 (solid
 554 lines, location of spectral extraction shown with arrows on panel (a)) and point spectra obtained with the point
 555 spectrometer on a large part of the sample surface (dotted lines, see Fig. 1 for measurement location), compared
 556 with laboratory reflectance spectra of known minerals taken from the PDS Geoscience Spectral Library. Laboratory
 557 spectra IDs are as follows: forsterite C3PO51, pigeonite C1PP42 and augite C1SC35. The silver resin spectrum was

558 measured outside of the meteorite, in the resin sample holder. Dashed lines refer to the absorption bands of another
 559 type of resin. Olivine, LCP and HCP are detected in the sample spectra, with overprinting of two types of resin
 560 signature.

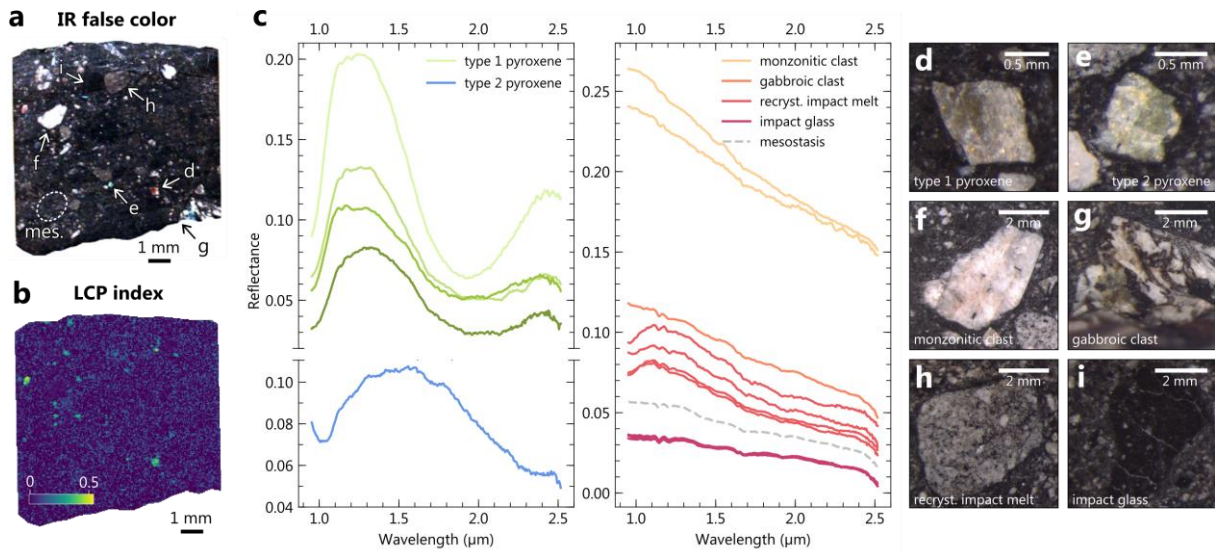
561



562

563 **Figure 8.** Hyperspectral cube of chassignite NWA 2737. **(a)** False RGB color composite in the near-infrared (R =
 564 1.22 μm, G = 1.66 μm, B = 2.15 μm) and associated RGB spectral parameters map below (R = olivine index, G = LCP
 565 index, B = HCP index). In the IR false color composite chosen, olivine, LCP and HCP appear blue, red/pink and
 566 green/yellow respectively. **(b)** Spectra extracted from the hyperspectral cube of NWA 2737 (solid lines, location of
 567 spectral extraction shown with arrows on panel (a)) and point spectra obtained with the point spectrometer on a
 568 large part of the sample surface (dotted lines, see Fig. 1 for measurement location), compared with laboratory
 569 reflectance spectra of known minerals taken from the PDS Geoscience Spectral Library. Laboratory spectra IDs are
 570 as follows: forsterite C3PO51, pigeonite C1PP42, augite C1SC35 and Fe/Ni alloy C1SC12. Resin spectrum was
 571 measured outside of the meteorite, in the resin sample holder. Dashed lines refer to the absorption bands of resin.
 572 Olivine, LCP and HCP are detected in the sample spectra, with overprinting of the resin signature. The dark areas
 573 correspond to olivine crystals. Their reflectance signature (i.e., low reflectance values and a moderate red slope over
 574 the VNIR range) is consistent with the presence of Fe/Ni alloy previously reported in this sample (e.g., Pieters et al.,
 575 2008).

576



577

578 **Figure 9.** Hyperspectral cube of the polymict breccia NWA 7034. **(a)** False RGB color composite in the near-infrared
 579 (R = 1.22 μm, G = 1.66 μm, B = 2.15 μm). In the IR false color composite chosen, LCP and HCP appear red/pink and
 580 green/yellow respectively. **(b)** Associated LCP spectral parameter map. **(c)** Spectra extracted from the hyperspectral
 581 cube of NWA 7034. Pyroxene types are defined by their spectral signature. Associated types of clasts observed
 582 through a stereo microscope are shown in **(d, e, f, g, h, i)**. Apart from pyroxenes, the clasts in NWA 7034 are
 583 associated with a blue slope in the near-infrared range, similarly to the point measurements on large clasts and
 584 over the whole chip. Recrystallized impact melts are associated with a subtle absorption band at ~1 μm, in
 585 agreement with the presence of small embedded pyroxene crystals.

586

587 Most of the spectra acquired on the polymict breccia NWA 7034 are dominated by a blue
 588 spectral slope and are not exhibiting any absorption band in the near-infrared (Fig. 9). Spectra of various
 589 types of clasts big enough to be resolved are extracted: pyroxene crystals (Fig. 9d and e), monzonitic
 590 feldspathic clasts (Fig. 9f), a gabbroic clast (Fig. 9g), pockets of crystalline and grainy pyroxene-feldspar
 591 impact melt (Fig. 9h), and pockets of dark impact glass with embedded small crystals (Fig. 9i). While no
 592 clear spectral signature of pyroxene is isolated with point spectroscopy due to spot size limitation (see
 593 section 4.2), the majority of the pyroxene clasts have spectra consistent with LCP, and one clast
 594 consistent with HCP in the hyperspectral cube (Fig. 9b and c). Isolated felsic clasts show high reflectance
 595 levels while not exhibiting any particular absorption band (Fig. 9c). The mesostasis, dark impact glass
 596 and gabbroic clasts do not exhibit any clear absorption band, while the pyroxene-feldspar crystallized
 597 impact melt pockets show the subtle absorption of pyroxene at ~1 μm (Fig. 9c).

598 In the NWA 4468 cube, shallow absorptions at 1.91 μm, 2.31 μm and 2.45 μm are detected in
 599 association with olivine (Fig. 7). The same bands are observed for NWA 1950 in association with olivine
 600 grains and in association with some LCP in areas (Fig. 8). While a mixture with a phyllosilicate or
 601 carbonate phase is not excluded as they are likely to produce absorptions at 1.9 μm and 2.3 μm, the set
 602 of shallow bands observed are consistent with epoxy contaminants (Fig. 7 and Fig. 8). In the DaG 489
 603 meteorite (not mounted with resin), bright areas show absorptions at 1.39 μm, 1.92 μm, 2.31 μm and

604 2.40 μm , consistent with the presence of a Mg-phyllsilicate, the best match being a saponite smectite
605 (Fig. 6). While a hydrous carbonate can present absorptions at $\sim 1.4 \mu\text{m}$, $\sim 1.9 \mu\text{m}$ and $\sim 2.3 \mu\text{m}$, the
606 distinctive 2.4 μm absorption identified in DaG 489 spectra is typical of phyllsilicate, not carbonate. In
607 NWA 1068 (2) sample, the absorptions at $\sim 1.4 \mu\text{m}$ and $\sim 1.9 \mu\text{m}$ are associated with the bright veins
608 crosscutting the rock and melt pockets and veins, but no additional band useful for mineralogical
609 characterization are observed. No other shallow absorption bands are clearly identified in NWA 1068
610 (1), NWA 2737, SaU 008 and Tissint cubes.

611

612 **4.2 Results from point spectra**

613

614 *4.2.1 Absolute reflectance*

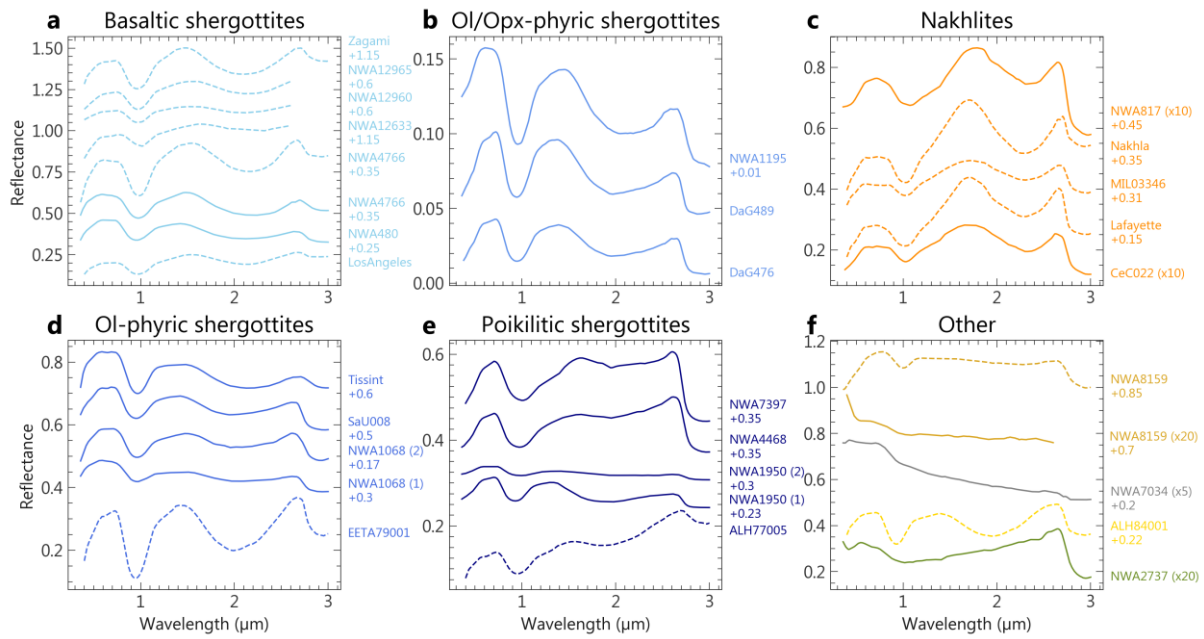
615 Martian meteorites are mostly dark in the VNIR range. At the standard observation geometry (ϕ
616 = 30° , $a = 0^\circ$), the mean maximum reflectance of chips/cut samples is 15%, and 95% of them do not
617 have a reflectance exceeding 25%. Such low reflectance values are expected for magmatic rocks with
618 low-silica content (e.g., Sgavetti et al., 2006). Powders are unsurprisingly more reflective with a mean
619 maximum reflectance of 34%, the most reflective sample being NWA 4766 (the maximum observed
620 reflectance is 57%). The darkest rocks from our suite are the nakhlite CeC 022 and the augite basalt NWA
621 8159, absorbing respectively at least 97% and 99% of the light in the explored spectral range. Finally,
622 powders exhibit redder spectra than rock samples (e.g., NWA 4766 in Fig. 10a).

623

624 *4.2.2 Band parameters related to main minerals*

625 Except for the polymict breccia NWA 7034, the chassignite NWA 2737 and the augite-basalt
626 NWA 8159, Martian meteorites reflectance spectra exhibit two broad absorption bands centered around
627 $\sim 1 \mu\text{m}$ and $\sim 2 \mu\text{m}$ (Fig. 10): bands I and II. These bands are explained by the combination of the
628 absorption bands of the main mafic minerals present in the meteorites: pyroxene and olivine (Fig. 2). In
629 addition, some spectra from the Martian meteorite suite exhibit shallow and narrow absorption bands
630 at $\sim 1.9 \mu\text{m}$ and $\sim 2.3 \mu\text{m}$, sometimes associated with an absorption at $\sim 1.4 \mu\text{m}$ and $\sim 2.45 \mu\text{m}$ (Fig. 11).

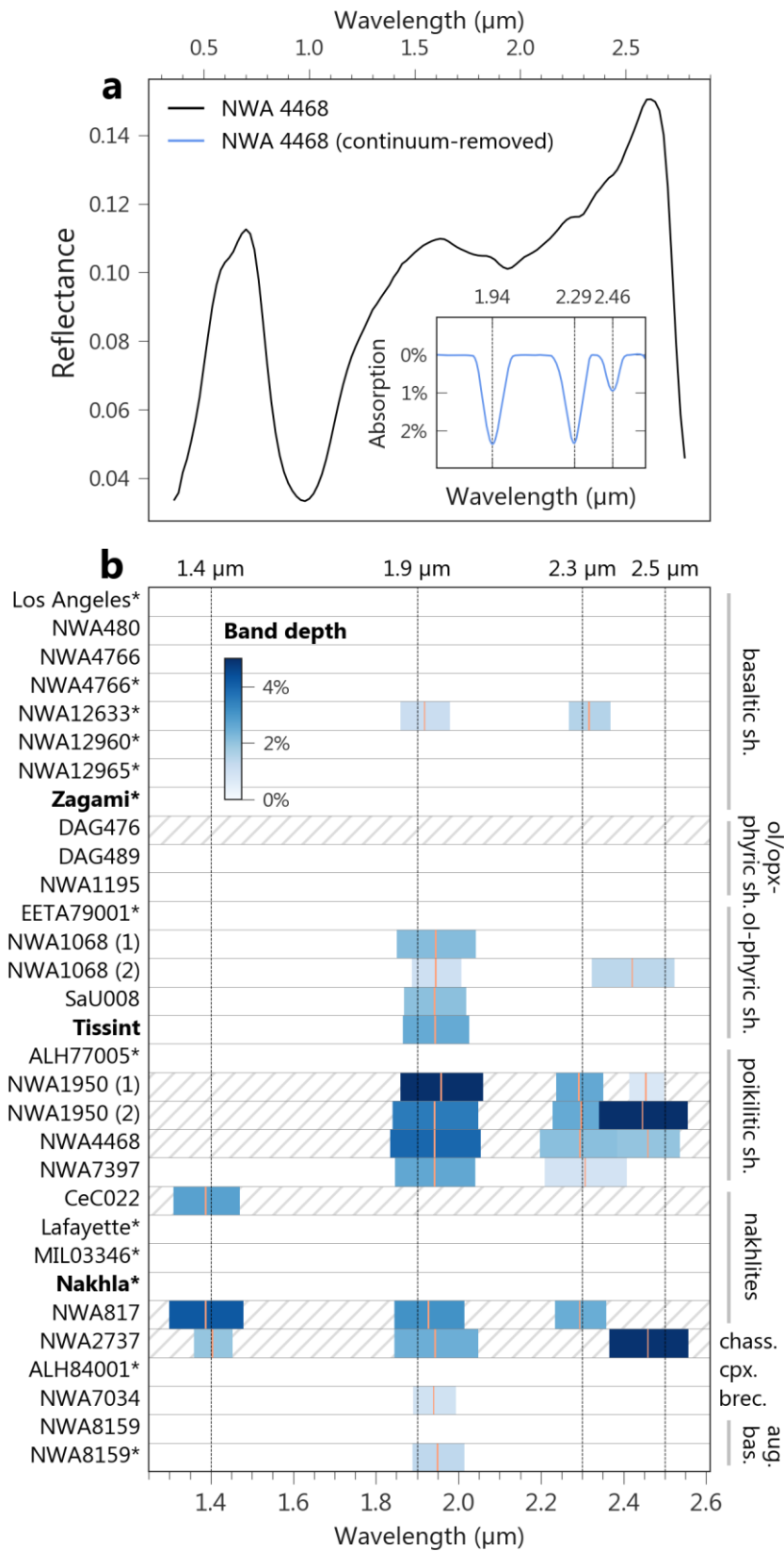
631



632

633 **Figure 10.** Spectra of the Martian meteorite suite obtained with the point spectrometer SHADOWS. Dashed and
 634 solid spectra respectively correspond to powder and chip/cut section samples. The spectra of CeC 022, NWA 817,
 635 NWA 2737, NWA 7034 and NWA 8159 are amplified in reflectance for clarity. The location of spot measurements
 636 on chip/cut section samples is given in Fig. 1.

637



638

639 **Figure 11. (a)** Point spectrum of shergottite NWA 4468 showing evidence of absorption bands at 1.95 μm , 2.30 μm
 640 and 2.45 μm . The continuum-removed spectrum is obtained by fitting and removing the 2 μm band of pyroxene.
 641 **(b)** Position and strength of the narrow absorption bands present in the meteorite point spectrum in the near-

642 infrared range. The band centers are indicated in blue. Falls are shown in bold and powders are indicated by an
643 asterisk sign. Samples mounted on epoxy (which might present absorptions in this wavelength range) are shown
644 with hatching.

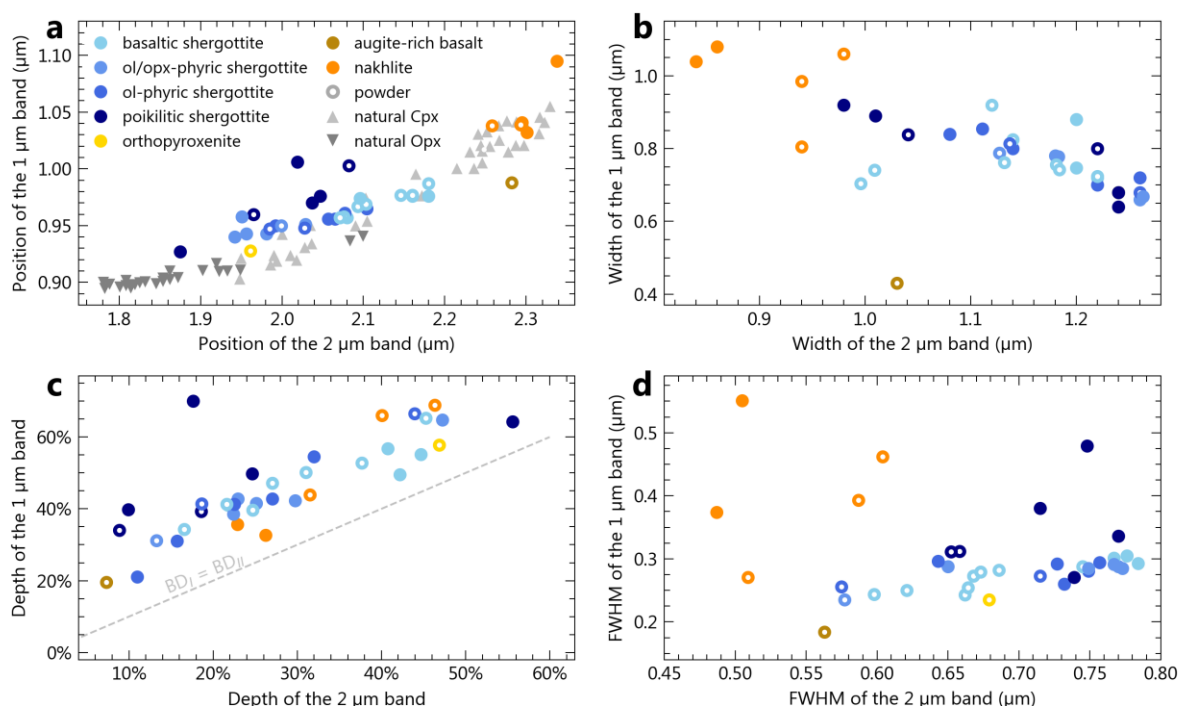
645
646 When mixed with pyroxene (usually the dominant phase in Martian meteorites), plagioclase or
647 maskelynite can be indirectly detected by a decrease of the pyroxene band depths and an increase of
648 the absolute value of reflectance at the band shoulders (Crown and Pieters, 1987). In our dataset,
649 flattening of the reflectance peak of pyroxene between band I and band II is observed in the shergottite
650 spectra, resulting in a particularly broad band I (e.g., NWA 1068 (2); Fig. 10). A similar feature was
651 commonly observed on previously measured shergottite spectra; however, applications of the Modified
652 Gaussian Method (MGM) by Sunshine et al. (1993) and McFadden and Cline (2005) showed that the
653 combination of low and high-calcium pyroxenes spectra solely could account for this effect. Hence,
654 flattening of the spectral shoulder of pyroxene in our dataset cannot be used as a means to infer a
655 plagioclase/maskelynite spectral detection. In addition, no correlation between the documented
656 proportion of maskelynite in the shergottites and the reflectance of the pyroxene shoulders or the depth
657 of the absorption is observed.

658 We report the position, depth, width and FWHM of band I and II of the meteorites' spectra in
659 Figure 12. The absorption position centers are compared with the ones measured on various natural
660 pyroxenes by Adams et al. (1974). Consistently with the prevalent mineralogy of the meteorites,
661 shergottites and nakhlites align well on the trend of clinopyroxenes, whereas ALH 84001 is closer to
662 orthopyroxenes (Fig. 12a).

663 Nakhlites are well distinguished from shergottites in the position of band I versus position of
664 band II diagram (Fig. 12a): position of band I is higher than $\sim 1.02 \mu\text{m}$ and position of band II higher than
665 $\sim 2.25 \mu\text{m}$. This is in line with the pyroxene Ca content: nakhlites dominant mineral, augite, is Ca-rich,
666 hence the absorptions centered at longer wavelengths. Nakhlites spectra are also associated with
667 specific band widths: band I is usually wider than $0.8 \mu\text{m}$, which is the case for some shergottites. This is
668 in line with their petrology: in nakhlites, pyroxene is primarily augite, which exhibits a band I wider than
669 the other pyroxene types (Fig. 2). However, band II is narrower for nakhlites than for other meteorites,
670 with a width smaller than $\sim 1 \mu\text{m}$, and a FWHM smaller than $\sim 0.6 \mu\text{m}$ (Fig. 12). We did not find
671 absorptions of alteration minerals in the spectra of nakhlites.

672 In shergottite spectra, the position of band I is lower than $\sim 1 \mu\text{m}$ and the position of band II
673 lower than $\sim 2.2 \mu\text{m}$ (Fig. 12a). Band I is usually slightly narrower than in the case of nakhlites (narrower
674 than $0.98 \mu\text{m}$), while band II is generally broader (larger than $0.98 \mu\text{m}$, with a FWHM superior to ~ 0.57
675 μm). The different types of shergottites have distinct spectral characteristics, though they are subtler
676 than between nakhlites and shergottites. Basaltic shergottites fall on the trend of natural pyroxenes from
677 Adams et al. (1974) and have band I and band II centered after $\sim 0.95 \mu\text{m}$ and $\sim 2.07 \mu\text{m}$ respectively (Fig.
678 12a). Shergottites with olivine are not perfectly aligned on the pyroxene trend, with the position of band
679 I over the position of band II ratio as well as the band I depth over band II depth II ratio increasing with
680 the olivine content (Fig. 12a and 12c). This is clearly seen in the poikilitic ("lherzolithic") shergottites, which

681 have the highest olivine content and a deeper band I, whose center is closer to their band II, compared
 682 to other shergottites (Fig. 12a). Spectra of olivine-bearing shergottites display band I at longer
 683 wavelengths, which is consistent with the relative position of the 1 μm band of olivine and the band I of
 684 low-Ca pyroxene. Olivine and olivine/orthopyroxene-phyric shergottites have a position of band I over
 685 position of band II ratio intermediate between those of poikilitic shergottites and natural pyroxenes, and
 686 share a band I and a band II centered below $\sim 0.95 \mu\text{m}$ and $\sim 2.1 \mu\text{m}$ respectively (Fig. 12a). These two
 687 subgroups are hardly distinguished from one another based on their band I and II. The augite basalt
 688 NWA 8159 exhibits faint absorptions at $0.98 \mu\text{m}$ (intermediate between those of shergottites and
 689 nakhilites) as well at $2.28 \mu\text{m}$ (in the nakhilites values range).
 690



691
 692 **Figure 12.** Parameters of the meteorite spectra corresponding to the two absorption bands of pyroxene around 1
 693 μm and 2 μm . Natural pyroxenes were measured by Adams et al. (1974). Other Martian meteorites whose
 694 absorptions at 1 μm and 2 μm were measured in previous studies were added to the plots: QUE* 94201, NWA
 695 6963, Shergotty (Filiberto et al., 2018), DaG 670, NWA 2626, NWA 6234, NWA 6963 and Y† 984028 (RELAB spectral
 696 library; all of these samples are shergottites).

697 * Queen Alexandra Range

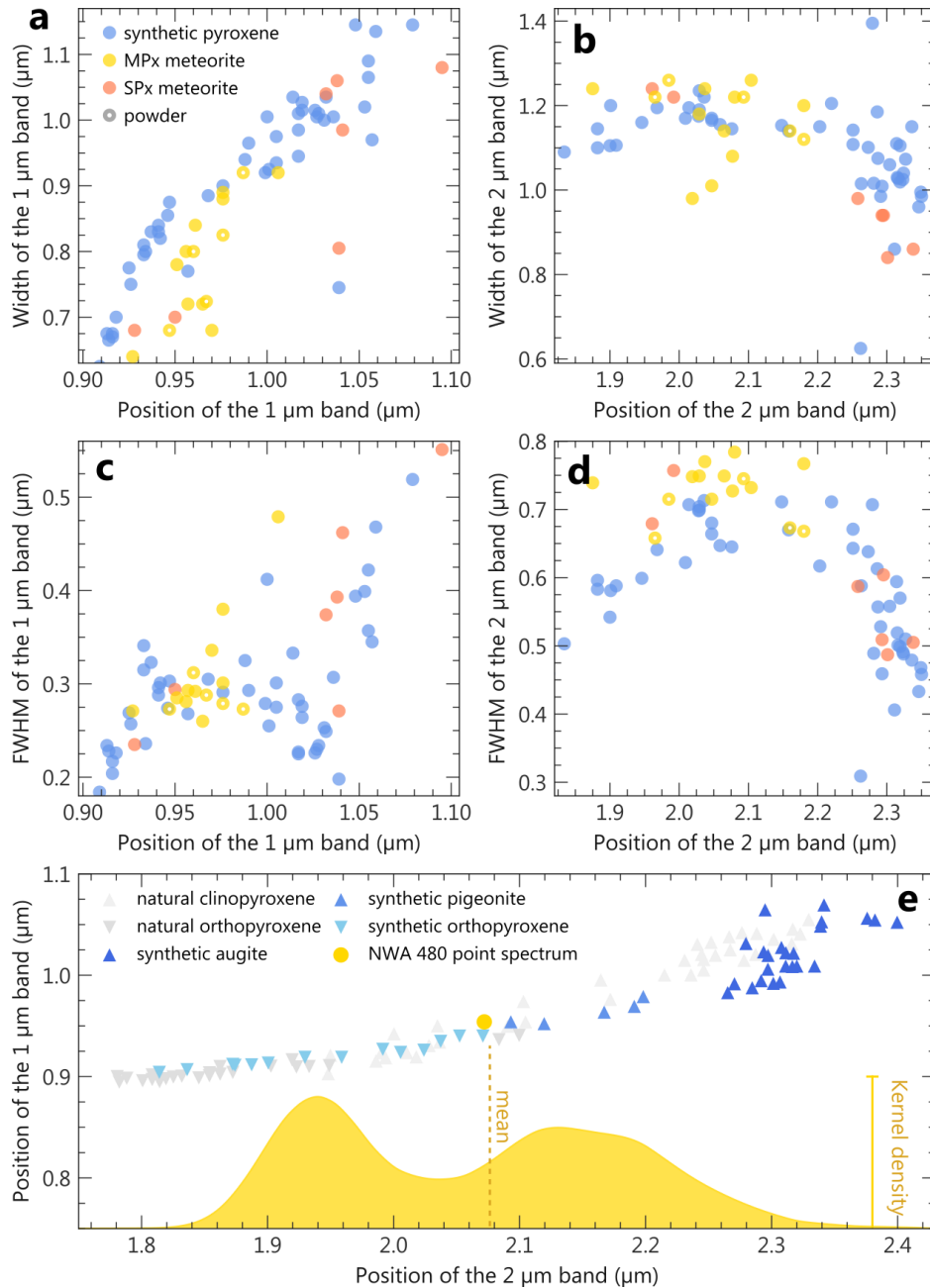
698 † Yamato

699

700 4.2.3 Discrimination between single pyroxene and multiple pyroxenes-bearing rocks

701 Based solely on the absorption band positions, a pyroxene of intermediate calcium content is
 702 spectrally hardly distinguishable from a mixture of low- and high-calcium pyroxenes. For instance, the
 703 point spectrum of shergottite NWA 480 shows a band II typical of a pyroxene of intermediate calcium

704 composition, while being actually composed of low and high-calcium pyroxenes (Fig. 13e). It has
705 previously been proposed that band width could be used to aid in the determination between pure
706 pyroxene and pyroxene mixture (e.g., Klima et al., 2007), as the superposition of LCP and HCP spectra
707 would result in wider absorptions. Figure 13 shows a comparison between band width and FWHM of
708 pure pyroxene spectra and spectra of measured meteorites – both on meteorites with multiple pyroxene
709 mineralogy (i.e., LCP and HCP) and those dominated by a single pyroxene mineralogy (i.e., mostly LCP
710 or HCP). These sets of measurements align together on the same trends and are hardly distinguishable
711 based on the width or FWHM of the pyroxene absorption bands. While not discriminant, FWHM of the
712 2 μm band seems to be the best parameter to help distinguish mixture of pyroxenes and single-pyroxene
713 mineralogy, as most of synthetic pyroxenes exhibit a slightly narrower band II (Fig. 13d). Interestingly,
714 multiple pyroxenes meteorites spectra usually exhibit a band I narrower than those of synthetic
715 pyroxenes, which is unexpected (Fig. 13a).
716



717

718 **Figure 13. (a, b, c, d)** Width and FWHM of the bands I and II as a function of band position for synthetic pyroxenes
 719 (Klima et al., 2007; 2011), compared to those of the point spectra of meteorites measured in this study. MPx refers
 720 to meteorites with multiple pyroxene mineralogy (i.e., LCP and HCP) while SPx refers to meteorites dominated by a
 721 single pyroxene mineralogy (i.e., mostly LCP or HCP). **(e)** Band positions of NWA 480 point spectrum and cube
 722 spectra, compared to those of synthetic (Klima et al., 2007; 2011) and natural (Adams et al., 1974) pyroxenes spectra.

723

724 4.2.3 NWA 7034, the polymict breccia

725

726

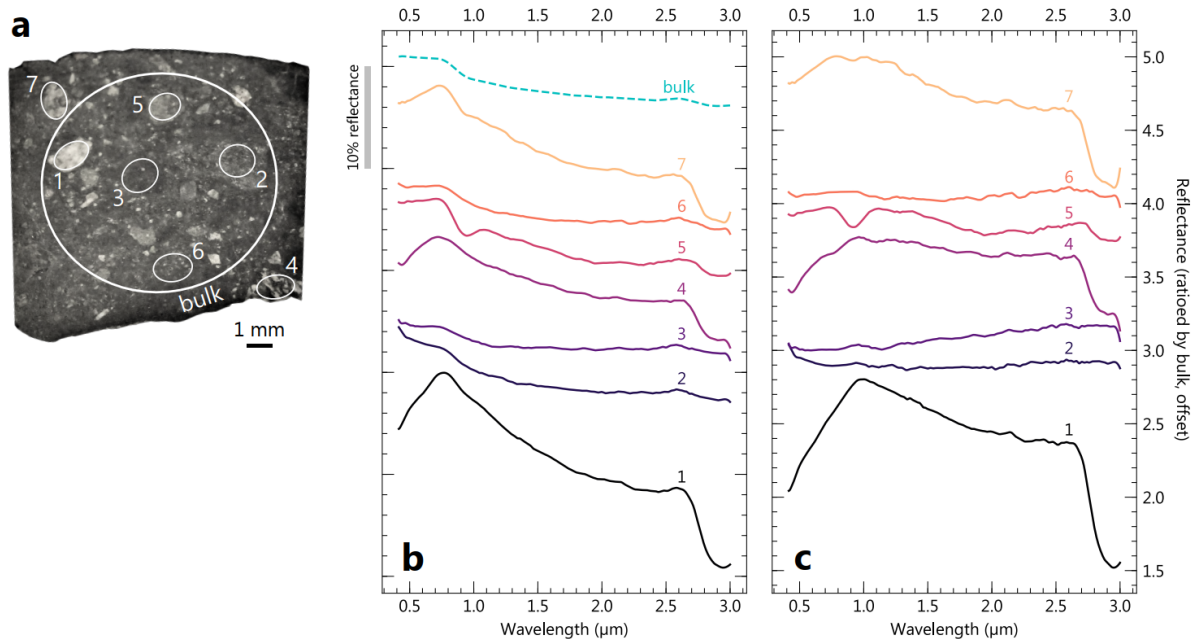
727

728

729

Point spectra acquired on a chip of NWA 7034 (polymict breccia) are consistent with previous VNIR measurements (Beck et al., 2015; Cannon et al., 2015) and show similarities with spectra from the hyperspectral cube: the sample is darker than most of the Martian meteorites from our suite, with a maximum reflectance of 11% in the visible and 7% at 2.5 μm (Fig. 10). This has been interpreted to be caused by the elevated content in magnetite and maghemite oxides (Beck et al., 2015). Whole chip and

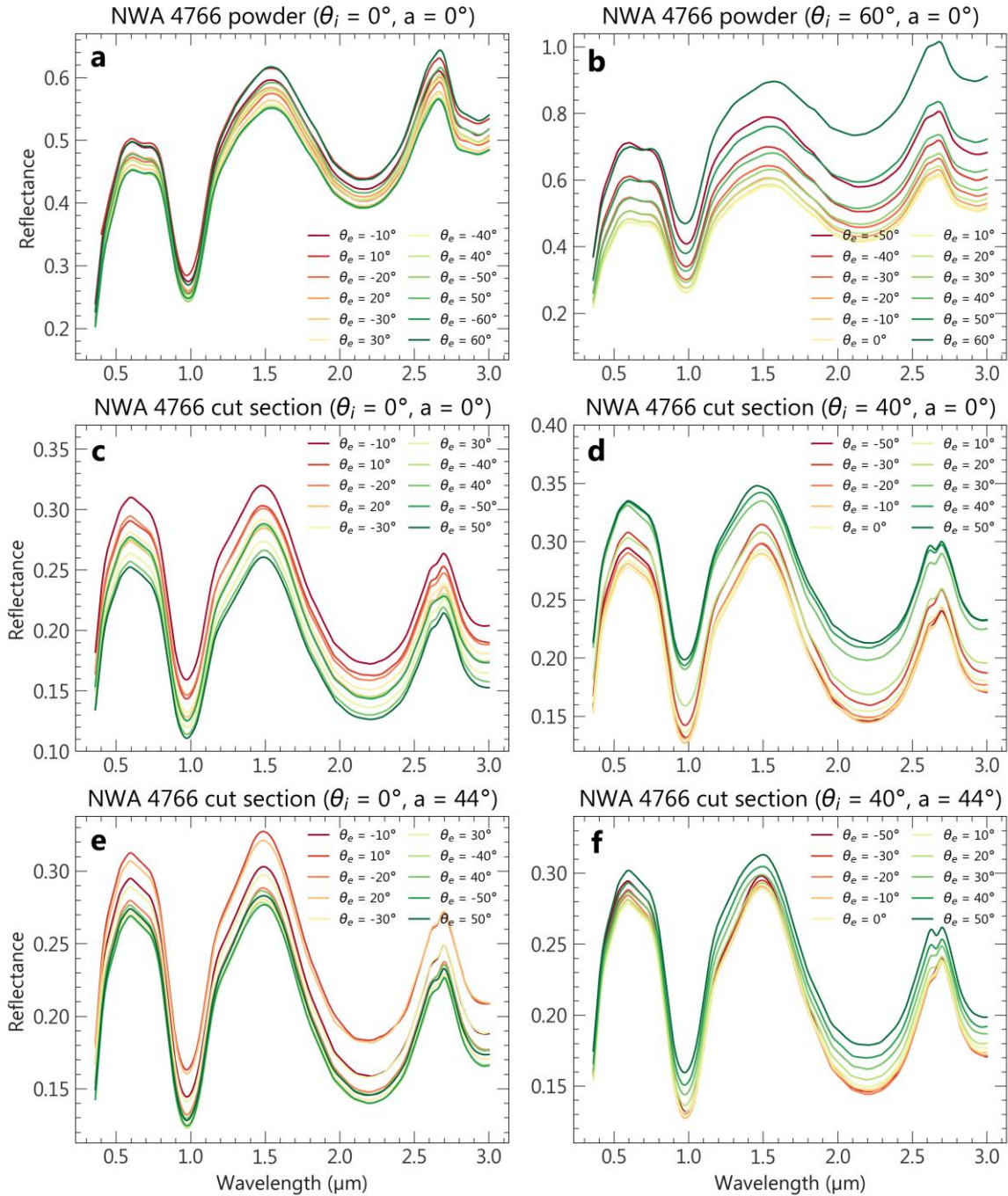
730 clasts spectra display a notable blue slope in the near-infrared (Fig. 14). Varying spectral slope as well
 731 as varying strength of the 3 μm hydration absorption band are observed from one clast to another.
 732 However, out of the seven clasts investigated, and apart from the 3 μm band, only one clast exhibits an
 733 absorption band in this spectral range (Fig. 14), showing that most of the breccia has spectral features
 734 dominated by the presence of oxides. This shallow band located at 0.95 μm is likely caused by the
 735 presence of a LCP clast in the measurement spot (Fig. 2). Limitation on the beam size prevented the
 736 analysis of smaller clasts, but further details are provided by imaging spectroscopy (section 4.1).
 737



738
 739 **Figure 14.** Reflectance spectra of NWA 7034 (polymict breccia). **(a)** Location of spot measurements on chip sample.
 740 **(b)** Spectra of various clasts are compared to the spectrum acquired over the whole cut section ("bulk"). **(c)** The
 741 same reflectance spectra are ratioed by the bulk spectrum.

742
 743 **4.3 Influence of the observation geometry**

744
 745 Bidirectional spectra acquired at various observational geometries on the particulate sample
 746 and cut section of NWA 4766 are shown in Figure 15. For both samples, variations of absolute reflectance
 747 and bands strength with changing observational geometries are observed.
 748



749

750 **Figure 15.** Spectra of NWA 4766 (basaltic shergottite) acquired with the spectro-gonio radiometer SHADOWS at
 751 various observational geometries, with emergence angles between -60° and 60° by steps of 10° for the powder and
 752 between -50° and 50° by steps of 10° for the cut section sample. Spectra are color-coded based on the absolute
 753 value of the phase angle (low: red, high: green). Technical limitations restrict measurements at low phase angles,
 754 hence spectra corresponding the following emergence angles are absent: **(a)** 0° , **(b)** -60° , **(c)** 0° , **(d)** -40° , **(e)** 0° , **(f)**
 755 -40° .

756

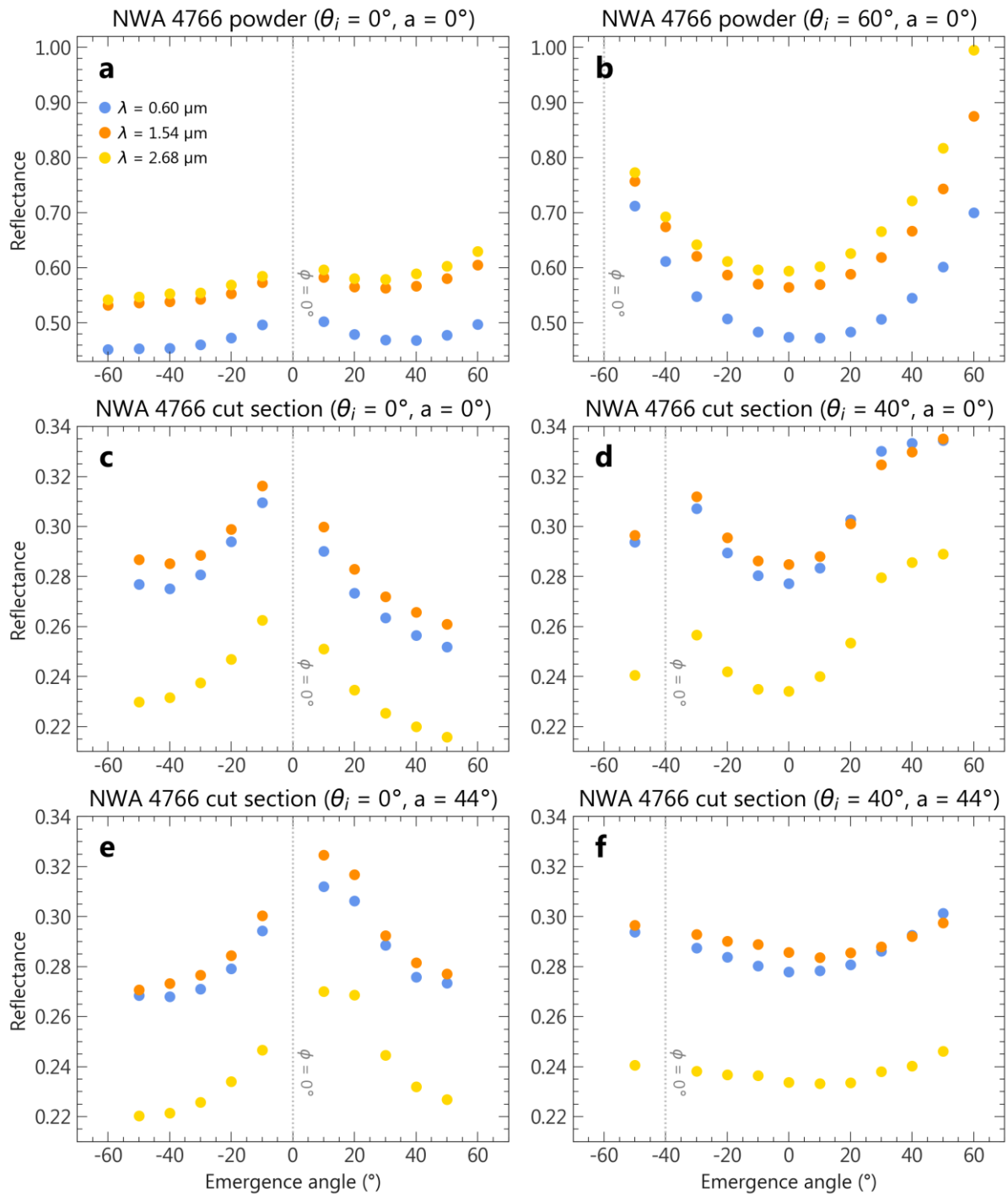
757 *4.3.1 Strength of the reflected light for varying emergence/phase and incidence angle*

758 The lowest reflectance levels are usually observed for phase angles of 40-70°, while the highest
759 are observed at phase angles close to 0°, as well as at the highest ones we were able to measure (90°
760 for the cut section and 120° for the powder; Fig. 16). Overall, both particulate and rock samples behave
761 as non-Lambertian surfaces and show some increase of their absolute reflectance at low phase angle,
762 indicative of backward scattering of the light by the sample. This effect is best depicted in BRDF
763 (Bidirectional Reflectance Distribution Function) polar diagrams, where the intensity of the reflected rays
764 at a given wavelength (here outside of the absorption bands of pyroxene) is shown as a function of their
765 emergence angle θ_e (Fig. 17). Since high phase angle measurements were limited, there are no data
766 points at ϕ higher than 90° and 120° for the cut section and powder, respectively. This means that any
767 forward scattering behavior is hardly explored through our measurements. However, measurements at
768 $\phi > 90^\circ$ on the powder show an increase of the reflected light intensity towards higher phase angle,
769 indicating that the sample does produce some forward scattering. The intensity of the backward
770 scattering, which we measured by ratioing the reflectance at $\theta_i = 0^\circ$, $\theta_e = 30^\circ$ by the reflectance at the
771 lowest phase angle measurement ($\theta_i = 0^\circ$, $\theta_e = 10^\circ$), is relatively similar for the particulate sample and
772 for the cut section (Fig. 16). At 600 nm, these ratios are equal to 1.07 and 1.10 for the powder and cut
773 section respectively. The same intensity of forward scattering is observed for measurements at ($\theta_i = 0^\circ$,
774 $\theta_e = -10^\circ$) and ($\theta_i = 0^\circ$, $\theta_e = -30^\circ$), despite the asymmetry in the reflectance level patterns (Fig. 16).

775 Continuum reddening of the powder spectra is seen to increase with the phase angle, at any
776 incidence. For instance, the reflectance at 2.68 μm at $\theta_i = 60^\circ$ and $\theta_e = 60^\circ$ ($\phi = 120^\circ$) is ~1.4 times the
777 reflectance at 0.60 μm , while it is ~1.1 times at $\theta_i = 60^\circ$ and $\theta_e = -50^\circ$ ($\phi = 10^\circ$). This effect is also observed
778 on the cut section spectra but is minor compared to the powder observed at the same geometry, with
779 up to 5% of reddening at $\phi = 90^\circ$ compared to $\phi = 10^\circ$.

780 At low phase angle, the intensity of the light reflected by the powder increases for oblique
781 incident light (i.e., high θ_i) compared to geometries where the incoming light source is at nadir (i.e., $\theta_i =$
782 0° ; Fig. 16 and Fig. 17), meaning that the sample tends to produce more backscattered light when
783 illuminated obliquely.

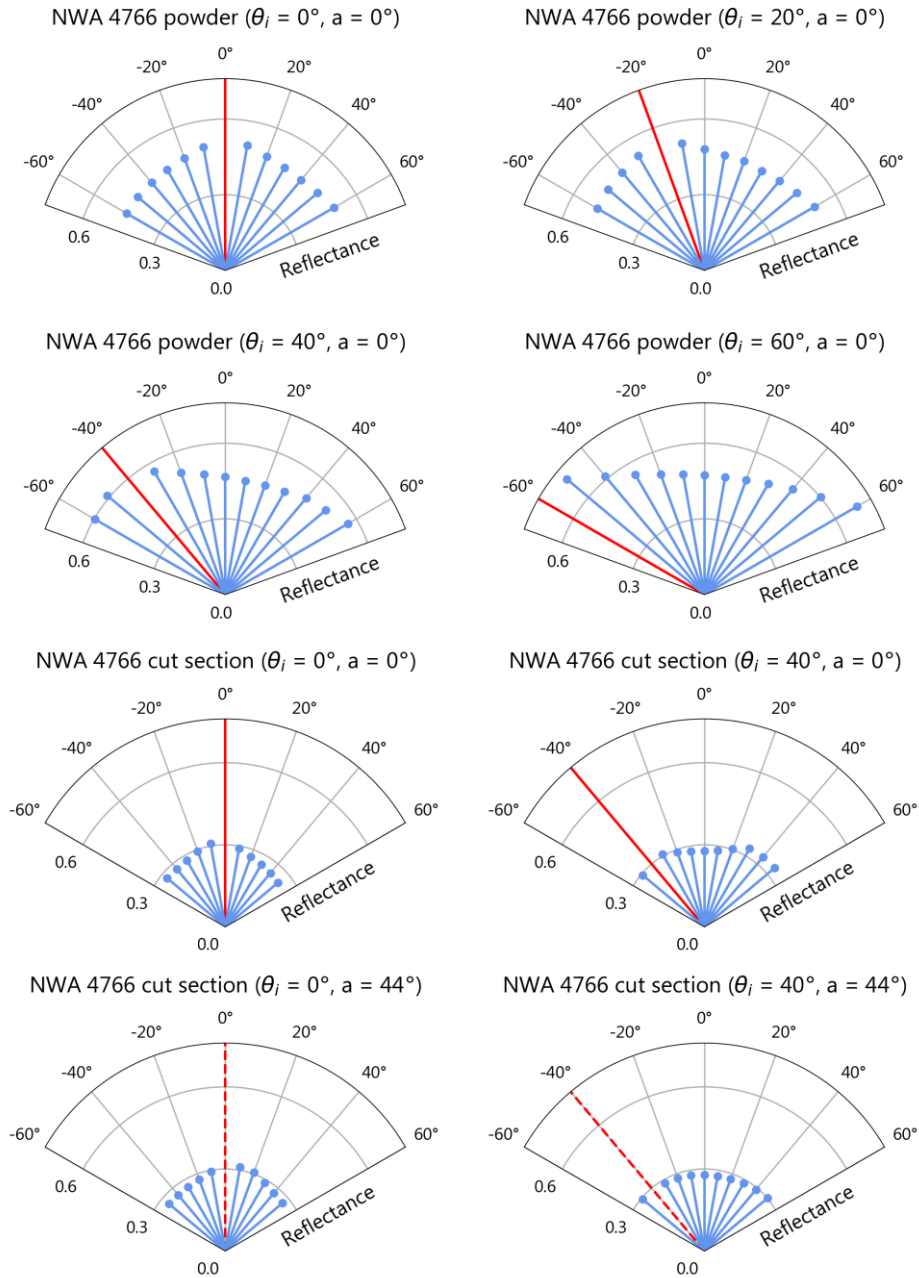
784



785

786 **Figure 16.** Reflectance of NWA 4766 (basaltic shergottite) obtained at various incidence, emergence and azimuth
 787 angles, with emergence angles between -60° and 60° by steps of 10° for the powder and between -50° and 50° by
 788 steps of 10° for the cut section sample. The reflectance was measured outside of the absorption bands of pyroxene,
 789 at the following wavelengths: $0.60\ \mu\text{m}$ (blue dots), $1.54\ \mu\text{m}$ (orange dots), $2.68\ \mu\text{m}$ (golden dots). Asymmetry in the
 790 reflectance levels pattern is explained by non-perfectly flat samples during measurement.

791



792

793 **Figure 17.** BRDF at $0.6\ \mu\text{m}$ for NWA 4766. The blue lines correspond to the reflectance for various emergence angles
 794 (see Table 1 for angle values). The incidence angle is indicated with a red line, dashed when the illumination is not
 795 in the principal plane ($a \neq 0^\circ$). Moderate backward scattering behavior is observed for both types of sample, while
 796 only the particulate sample shows clear moderate forward scattering behavior (at the observation geometries
 797 explored).

798

799 4.3.2 Reflected light strength outside of the principal plane

800 The effects of measuring the reflected light at varying azimuth angle on reflectance spectra can
 801 be assessed through bidirectional measurements on the cut sample, where the reflected light was
 802 measured at positive azimuth (Table 1). Observations with variable azimuth for vertical illumination seem

803 to generate similar spectra in terms of absolute reflectance (see Fig. 16c compared to 16e) and band
804 depths (see red curve compared to orange curve in Fig. 18d and 18e) than measurements in the principal
805 plane. At oblique incident light, changes in azimuth produce distinct spectra than those measured in
806 the principal plane: the absolute reflectance shows little variation at diverse emergence angles (Fig. 15,
807 Fig. 16, and Fig. 17), suggesting that forward scattering probably does not extend laterally outside of
808 the principal plane.

809 Overall, considering all the bidirectional spectra at various incidence, emergence and azimuth
810 angles, the maximal variation of reflectance observed at the same wavelength is 9% for the cut section
811 and 46% for the powder. This value for the powder drops to 14% when considering only the observation
812 geometries that were explored commonly for both sample types (the variation for the cut sample
813 remains unchanged). If the two types of samples and all geometries are considered, the maximum
814 reflectance variation spectrum-to-spectrum is up to 79% of reflectance for NWA 4766.

815

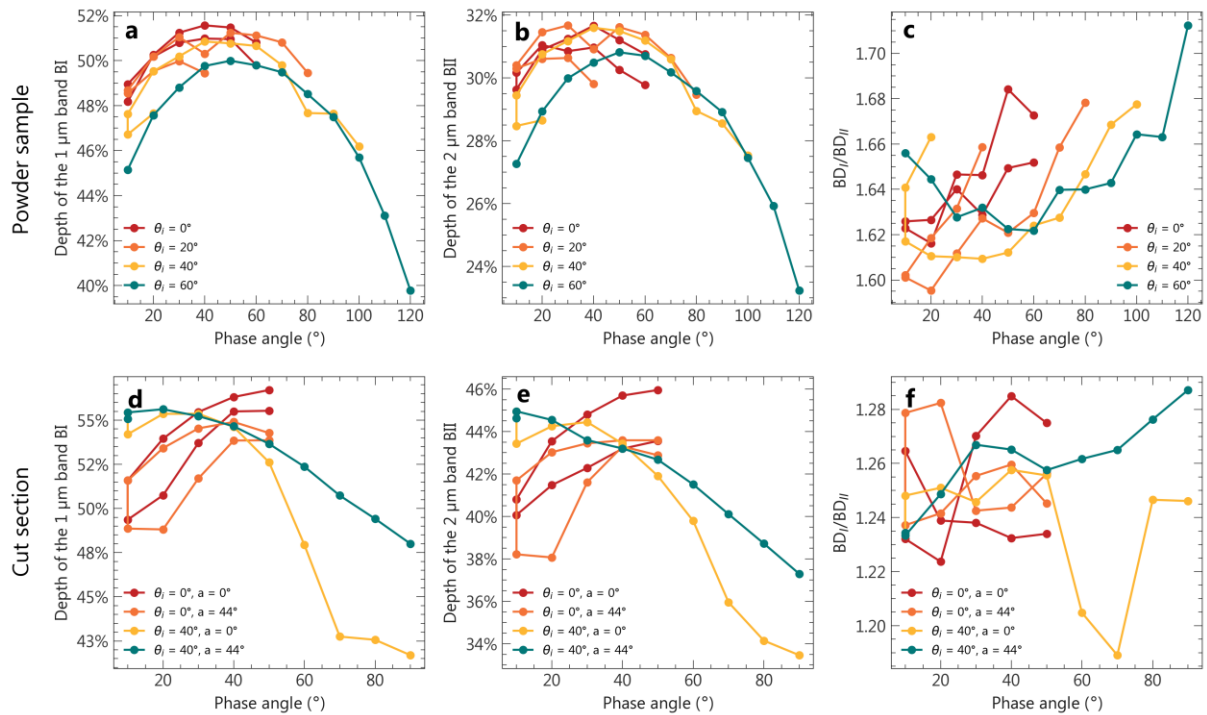
816 *4.3.3 Effect of the observational geometry on band parameters*

817 No significant shift in band positions is observed with varying observational geometry: no more
818 than 5 nm for band I and no more than 60 nm for band II. These little variations appear uncorrelated
819 from the reddening of the spectra and are most likely caused by uncertainties in continuum removal, or
820 slight variations of the grains measured between different observational geometries (as the
821 measurement spot varies).

822 The strength of band I and II show variations with changing observation angles, respectively up
823 to 12% and 9% for the particulate sample and up to 15% and 13% for the cut section. This leads to the
824 BD_I/BD_{II} ratio increasing with the phase angle in most cases, with up to 10% of variation given the
825 observational geometry. The variation of band depths depending on the observational geometry seems
826 to be anticorrelated with the variation of reflectance: maximum band depths for both absorption bands
827 are observed when the reflectance is low ($\phi \sim 40-60^\circ$), while the lowest band depths are observed at ϕ
828 close to 0° and at $\phi = 120^\circ$ which correspond to high absolute reflectance (Fig. 16 and 17). Similar to
829 what is observed for the absolute reflectance, band depths appear to fluctuate less at high incidence
830 and high azimuth (green curve in Fig. 18d and 18e), than in the principal plane at the same incidence
831 (yellow curve in Fig. 18d and 18e).

832 Considering both samples and all explored geometries, strength of band I and II vary by up to
833 17% and 23% respectively.

834



835

836 **Figure 18.** Variation of the pyroxene bands parameters as a function of the observational geometry, for the powder
 837 **(a, b, c)** and cut section **(d, e, f)** samples of NWA 4766.

838

839 **5 Discussion**

840

841 **5.1 Comparison of point spectra and cubes**

842

843 As expected, the point spectra acquired on the meteorites with the SHADOWS spectrometer
 844 result from the spectral mixing of various components. In NWA 480, three main phases are identified in
 845 the spectral imagery: the 2 μm absorption band centers indicative of low and high-calcium pyroxenes
 846 (from the core to the rim of the crystals), and a groundmass with an intermediate 2 μm band center and
 847 lower reflectance values. Comparison with SHADOWS measurement shows that these three components
 848 are spectrally mixed in the point spectrum of NWA 480, which exhibits intermediate 2 μm band center
 849 and moderate reflectance values (Fig. 5b). In ol-phyric and ol/opx-phyric shergottites cubes, olivine
 850 phenocrysts and dark areas embedded in a pyroxene groundmass with a dominant low-calcium
 851 signature are identified, while their point spectra are dominated by the spectral signature of the
 852 groundmass (Fig. 6b). Low reflectance value in the point spectra can be explained by the presence of
 853 the olivine phenocryst and the dark areas, which have lower reflectance in the hyperspectral cubes. The
 854 point spectra of the poikilitic shergottites also appear to be a spectral mixing of olivine and pyroxene
 855 signatures, with reflectance values intermediate between those of the olivine, LCP and HCP crystals (Fig.
 856 7b).

857 These results indicate that significant spectral mixing occurs in the point measurements at spot
 858 sizes down to a few millimeters, which is SuperCam’s spot size at a few meters from the target (Wiens

859 et al., 2017). At this resolution, imaging spectroscopy (i.e., MicrOmega's technique) is more powerful to
860 isolate the various main minerals and minor phases. However, our previous results on band parameters
861 show that point spectrometry is sufficient to discriminate the different Martian meteorites families and
862 to identify their main mineralogy.

863

864 **5.2 Nature of the alteration in Martian meteorites**

865

866 Alteration phases have been reported in many of the meteorites studied here (see dedicated
867 section 2.2). We show that some Martian meteorites have spectra with shallow absorption bands in the
868 near-infrared, which usually point to the presence of alteration phases. In particular, bands at $\sim 1.9 \mu\text{m}$,
869 $\sim 2.3 \mu\text{m}$ and $\sim 2.4\text{-}2.5 \mu\text{m}$, usually attributed to hydrated minerals are observed (Fig. 11). Unfortunately,
870 resin contaminants likely present in several of the samples and absorbing at these same wavelengths
871 prevent any definitive conclusion on the ability of reflectance spectrometry to identify these phases in
872 most of our Martian meteorites suite. Nonetheless, several other samples exhibit these bands while
873 never have been embedded in resin: DaG 489, NWA 1068 pairs, NWA 7034, NWA 7397, NWA 8159,
874 NWA 12633, SaU 008 and Tissint (Fig. 6 and 11).

875 Our measurements with imaging spectroscopy show that, in DaG 489, a smectite phase
876 occurring in patches in the sample is the cause of the shallow absorption bands observed in the point
877 spectrum. In paired DaG 476, the bright mineral phase occurring in fractures, in grain boundaries and
878 filling cavities was identified to be a Ca-carbonate (Greshake and Stoeffler, 1999; Zipfel et al., 2000;
879 Mikouchi et al., 2001). While carbonate, if present in our sample, is not detected in DaG 489 with imaging
880 spectroscopy, Greshake and Stoeffler (1999) and Mikouchi et al. (2001) reported iddingsite (mixture of
881 oxides, ferrihydrites and poorly crystalline phyllosilicates rich in iron and magnesium) associated with
882 altered olivine grains of DaG 476. The iddingsite is probably the phase detected here, and in particular
883 the phyllosilicate component, which we identify to be a Mg-smectite (a common product of olivine
884 alteration, e.g., Dehouck et al., 2014).

885 Similar to the Dar al Gani pair, the terrestrial Ca-carbonate phase which fills fractures in the NWA
886 1068 pair (Barrat et al., 2002) was not detected in the point spectra or in the hyperspectral cubes. Instead,
887 these white fractures have the spectral signature of hydration, with bands at $\sim 1.4 \mu\text{m}$ and $\sim 1.9 \mu\text{m}$, and
888 lack the carbonate bands at $\sim 2.3 \mu\text{m}$ and $\sim 2.5 \mu\text{m}$. Hydrous carbonates can have similar features with
889 subdued $2.3 \mu\text{m}$ and $2.5 \mu\text{m}$ (Harner et al., 2015) and, although more common in cold desert finds, were
890 reported in hot desert finds (Miyamoto, 1991). However, hydrous Ca-carbonate such as
891 monohydrocalcite are usually metastable with respect to calcite and aragonite (Hull and Turnbull, 1973)
892 and are not expected to precipitate extensively in a hot desert environment, or to be preserved during
893 a long-term storage at room temperature. Hydrous Mg-carbonates such as hydromagnesite or
894 nesquehonite on the other hand, can be stable if the formation of magnesite (anhydrous Mg-carbonate)
895 is kinetically inhibited (i.e., by temperature or CO_2 partial pressure), and have been reported in weathered
896 chondrites (Marvin and Motylewski, 1980; Hänchen et al., 2008). Consequently, a terrestrial hydrous Mg-
897 carbonate might be the phase detected in the NWA 1068 pair.

898 The alteration phases causing the shallow absorptions in the other meteorites without resin are
899 quite challenging to determine, as hyperspectral imaging of the samples fails at isolating these phases
900 to retrieve interpretable spectra, at least at the spatial resolution and SNR of our measurements. Of
901 particular interest is the 1.9 μm band identified in Tissint spectrum; as this meteorite is an observed fall,
902 any hydrated mineral is likely of Martian origin. The only mineral that can exhibit hydration and that has
903 been documented in Tissint is apatite, which is reported in minor phases (less than 1%; Chennaoui
904 Aoudjehane et al., 2012). The absorption in Tissint spectrum is very subtle ($\sim 1\%$ of absorption after
905 removal of the pyroxene 2 μm band), which is agreement with the very low content of hydrated minerals
906 reported. Also, water adsorption during the measurement cannot be excluded.

907

908 **5.3 Observational geometry considerations**

909

910 As expected, the Martian sample studied both in powder and section form (NWA 4766) shows
911 a non-Lambertian behavior with significant backward scattering. A forward scattering peak is observed
912 for the powder sample, while it could not be investigated on the chip sample (as no measurement at a
913 phase angle higher than 90° was performed).

914 Notably, an anticorrelation between reflectance and band depth was measured: at observational
915 geometries minimizing the reflectance level (usually for $\phi \sim 40\text{--}60^\circ$), maximization of band depth was
916 observed, and vice versa. Pommerol and Schmitt (2008), Shepard and Cloutis (2011) and Beck et al.
917 (2012) observed the same trend at high phase angle, respectively on a smectite and volcanic tuff, on
918 lazurite and on meteorites. Pommerol and Schmitt (2008) proposed that, for these observational
919 geometries, the proportion of photons scattered by direct reflections on the surface grains compared
920 to the absorbed photons is more important, causing a decrease of band strength at high phase angle.
921 This arch shape (Fig. 18) observed for the evolution of band depths as a function of phase angle was
922 observed by Beck et al. (2012) and Schröder et al. (2014) with reflectance ratios and is likely linked with
923 the spectral slope, though band depths formulation is more complex than reflectance ratios.

924 The increase of reddening with phase angle in the powder's spectra was also observed by Beck
925 et al. (2012) and Schröder et al. (2014), where some microscopic roughness effects, unpredicted by the
926 classical radiative models, are proposed to explain this spectral behavior.

927 The SuperCam visible and infrared spectrometers acquire spectra at different sol hours, which
928 makes the phase angle and azimuth between the Sun (light source) and the rover's Mast Unit (which
929 collects the reflected light) highly variable from one observation to another (Fouchet et al., submitted).
930 Previous bidirectional measurements have already shown the significant impact of a changing
931 observational geometry on the spectral reflectance levels and band strengths. Hence, the main goal of
932 SuperCam reflectance spectrometers will be the mineralogical identification rather than the
933 quantification, which can be biased if relying on band depth. We quantified the significance of these
934 changes on a Martian sample for phase angles ranging from 10° to 120° and for positive azimuth.
935 Though the rocks that will be analyzed by the Perseverance rover will be very diverse, and will include
936 sedimentary and igneous rocks (e.g., Goudge et al., 2015; 2017; Horgan et al., 2020), our results illustrate

937 the expected variations in reflectance and band depths due solely to changes in the observation
938 geometry.

939 For our sample and angles explored, the absolute reflectance of the powder can be doubled,
940 while variations of only ~10% are observed for the rock sample. Thus, while caution should be applied
941 when interpreting future measurements from SuperCam for the reasons developed above, it is noted
942 that the effects of variable observation geometries on the absolute reflectance are less important for
943 the rock than for the loose material. In addition, while the powder is remarkably more reflective than
944 the rock slab, pyroxene band strengths are comparable in spectra of both samples. Of interest for the
945 identification of subtle bands associated to alteration minerals, there is a band strength optimum
946 depending on the phase angle (in our case between 40° and 60°). However, this optimum also
947 corresponds to minimal absolute reflectance, hence lower SNR measurements. A phase angle
948 compromise between absolute reflectance and band strength is recommended. Measurements
949 performed at angle $\pm\phi$ around this optimum are expected to be comparable both in terms of reflectance
950 level and band depth. Additionally, measurements performed with different azimuths are also expected
951 to be comparable in terms of absolute reflectance and band depths if they are acquired close to the
952 nadir – if ever achieved on Mars.

953

954 **5.4 Implications for the spectral study of igneous rocks on Mars**

955

956 Our sample suite reflects the current global diversity of Martian meteorites, where effusive rocks
957 are rare and intrusive rocks dominant. Thus, the spectral features of most of our samples should not be
958 directly compared to the orbital observations of the surface – except from outcrops where rocks have
959 been excavated from depth, by erosional processes or during an impact (e.g., rocks in central peaks of
960 complex craters; Brustel et al., 2019). Nevertheless, this case study can be interpreted in terms of phase
961 detectability and mixing in the spectral data.

962 Over the last decades, reflectance spectroscopy between ~1 μm and ~2.6 μm has been used to
963 study the diversity of the Martian crust at a global scale (e.g., Bibring et al., 2005; Mustard et al., 2005).
964 The predominant detection of pyroxene (and olivine) indicates that the crust is dominated by poorly
965 evolved lithologies. The detection of felsic rocks by reflectance and thermal infrared measurements over
966 the highlands (Christensen et al., 2005; Bandfield et al., 2006; Carter and Poulet, 2013; Wray et al., 2013),
967 in situ at Gale crater (Sautter et al., 2015) and in the polymict breccia NWA 7034 and pairings (Agee et
968 al., 2013) showed that at least locally, evolved lithologies exist on Mars. While feldspars are more
969 challenging to detect than pyroxenes when mixed with other minerals (Crown and Pieters, 1987), our
970 measurements on meteorites at high resolution show that, even isolated from the pyroxenes (i.e., in
971 spectral cubes), they are hardly detectable. High shock pressure during the impact(s) is likely to decrease
972 the ~1.3 μm absorption band depth of feldspars (Johnson and Hörz, 2003) but does not alone explain
973 the complete absence of absorption in our spectral data. Other explanations on the non-detection of
974 this band include low-iron content (as the absorption is caused by Fe^{2+}) or the presence of darkening
975 impurities in the feldspars. Felsic clasts in NWA 7034 are broadly featureless in the VNIR range: they

976 sample a primitive Martian crust that is probably undetectable from the orbit with reflectance
977 spectroscopy.

978 Important interpretations about the magmatic evolution and cooling history of the Martian crust
979 are derived from the types of pyroxene detected from the orbit (e.g., Baratoux et al., 2013). We show
980 that, based on spectral parameters, Martian meteorites with both low- and high-calcium pyroxene are
981 hardly distinguishable from rocks with a single intermediate type of pyroxene, while these rocks can be
982 formed via different magmatic processes (e.g., the presence of exsolution lamellae of new composition
983 can occur with slow cooling). We highlight the necessity to consider spectral mixtures when interpreting
984 pyroxene-dominated spectra, where deconvolution methods like the Modified Gaussian Models (MGM)
985 should be applied (Sunshine and Pieters, 1993). Nonlinear unmixing approaches such as the Hapke
986 (Hapke, 1981) or Shkuratov (Shkuratov, 1999) models might also address this issue, but they were
987 developed for particulate materials where grains are separated. Their applicability to compact rocks
988 remains to be assessed and validated.

989

990 **6 Summary**

991

992 By collecting an extensive suite of samples representative of the current Martian meteorites
993 diversity, we were able to produce spectral measurements of several samples of each family in the VNIR
994 range. The key findings are summarized below.

- 995 • The measurement spot of the spectro-gonio radiometer SHADOWS is analogous to the size of
996 those of Mars 2020 SuperCam VNIR spectrometer at ~2–4 m distance (Wiens et al., 2017). Overall,
997 our measurements with SHADOWS are able to recover the primary mineralogy of the Martian
998 meteorites suite, based on the absorption bands parameters: olivine, low and high-calcium
999 pyroxene. The presence of maskelynitized plagioclase is not clearly detected. Although not abundant
1000 in Martian meteorites, some hydrous phases are detected, but their identification is impeded by
1001 the spot size and is eventually achieved using high-resolution imaging spectroscopy (MicrOmega's
1002 technique). The alteration phases identified likely consist of a hydrous Mg-carbonate of terrestrial
1003 origin (in NWA 1068) and a Mg-smectite (in DaG 489).
- 1004 • The most effective distinction within Martian meteorites based on the point spectra is achieved
1005 using the positions of band I versus position of band II criterion, which enables the identification
1006 of nakhlites over shergottites, having different pyroxene composition and olivine contents.
1007 Similarly, further discrimination can be made inside the various shergottite classes, between
1008 basaltic, phryic and poikilitic shergottites. No correlation between spectral properties and ejection
1009 ages is observed.
- 1010 • At 2–4 m from the outcrop, the point spectra acquired by SuperCam are likely to suffer from
1011 spectral mixing. Here, and as confirmed by imaging spectroscopy, the point spectra acquired by
1012 SHADOWS are the result of a mixing between – when present – high and low-calcium pyroxenes,
1013 olivine and additional darkening phases (e.g., melt, oxides or alloys). Rocks dominated by a single
1014 type of pyroxene with intermediate calcium content and rocks dominated by both LCP and HCP

1015 cannot be easily distinguished from each other based on band parameters. However, using the
1016 FWHM of band II seems to help in the discrimination, being more often higher in the case of
1017 mixtures.

1018 • Multiple bidirectional measurements on a shergottite, the most abundant type of Martian
1019 meteorites, confirm their non-Lambertian behavior in both particulate and consolidated form, with
1020 forward scattering (at least for the powder) and backward scattering of the VNIR light. While
1021 strongly variable reflectance levels are observed for the powder with varying observational
1022 geometry, these variations are lower for the consolidated sample. The pyroxene absorption
1023 strengths show variations up to ~10–15%, comparable in both types of sample.

1024 • Of interest for in situ identification of minor minerals, the absorption strengths are maximal at
1025 moderate phase angle (here between 40° and 60°). Measurements performed outside of the
1026 principal plane and/or at angle $\pm\phi$ around this optimum are expected to be comparable both in
1027 terms of reflectance level and band depth.

1028

1029 The meteorite spectra are available for further studies in the supplementary materials.

1030

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