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5 **A multi-methodological study of the bastnäsite-synchysite polysomatic series: Tips and tricks of**  
6 **polysome identification and the origin of syntactic intergrowths**

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21

22 **Abstract**

23

24 In this paper, we evaluated the potentialities of Raman spectroscopy and electron backscattered  
25 diffraction (EBSD) in the microscopic characterization of Ca-REE fluorcarbonates (CRFC) belonging  
26 to the bastnäsite-synchysite series to provide a “road map” for further investigations with transmission  
27 electron microscopy (TEM). EBSD was effective in establishing the sample orientation, setting up the  
28 oriented cuts, and ascertaining the effective syntactic relationship among all the detected CRFC phases;  
29 however, it failed to distinguish between different polysomes. On samples with different orientations  
30 that were preventively ascertained by EBSD and characterized by scanning electron microscopy (SEM)  
31 coupled with energy dispersive X-ray spectroscopy (EDS), performing micro-Raman spectroscopy  
32 allows distinguishing between polysomes based on the differences in intensity and position of the  
33 symmetric stretching vibration ( $\nu_1$ ) of the carbonate group ( $\text{CO}_3^{2-}$ ) in the region around 1080–1099  $\text{cm}^{-1}$   
34 <sup>1</sup>. However, as evidenced by TEM-EDS, what appears as a homogeneous polysome in backscattered  
35 electrons (BSE) images may be a disordered intergrowth of compositional faults with a bulk

36 composition being matched with that of a real polysome only by accident. Therefore, we conclude that  
37 the Raman signal is sensitive to different Ca/(Ca+REE) ratios but not to any ordered distribution of Ca-  
38 poor and Ca-rich lamellae within the analyzed volume, making the unambiguous identification of a  
39 polysome tricky. Finally, several ordered polysomes were detected at the TEM scale, including a  $B_2S$   
40 and a long-range polytype with a 32 nm repeat distance along  $c$ . The possible implications of the  
41 detected microstructure for ore mineral formation are discussed.

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### Introduction

45 The Ca-REE fluorcarbonates (hereafter CRFC) are important minerals for at least two (apparently)  
46 distant reasons, one tied to fundamental research and the other to critical raw materials. Indeed, from a  
47 crystallographic point of view, CRFC form a polysomatic series (Veblen, 1991) with bastnäsite  
48  $[\text{REE}(\text{CO}_3)\text{F}]$  and synchysite  $[\text{CaREE}(\text{CO}_3)_2\text{F}]$  as the end members (Fig. 1). Accordingly, intermediate  
49 terms can be described by bastnäsite ( $B$ ) and synchysite ( $S$ ) modules ( $B_nS_m$ ) and their composition  
50 calculated as  $[\text{REE}(\text{CO}_3)\text{F}]_m \cdot [\text{CaREE}(\text{CO}_3)_2\text{F}]_n$  (Donnay & Donnay, 1953). Intermediate terms of the  
51 series are parisite  $[\text{CaREE}_2(\text{CO}_3)_3\text{F}_2]$  (or  $BS$ ) and röntgenite  $[\text{Ca}_2\text{REE}_3(\text{CO}_3)_5\text{F}_3]$  (or  $BS_2$ ). Possible  
52 additional intermediate polysomes have been described by high-resolution transmission electron  
53 microscopy (HRTEM) studies (van Landuyt & Amelinckx, 1975; Wu et al., 1998; Meng et al., 2001a,  
54 b, 2002; Ciobanu et al., 2017; Capitani, 2019, 2020; Zeug et al., 2021). Moreover, the layer sequence  
55 within a polysome may be different: a given layer ( $B$  or  $S$ ) may be differently rotated with respect to  
56 the ordered sequence, giving rise to polytypism and polytypic disorder as in micas (Banfield et al.,  
57 1994). Finally, within a polysome,  $B$  and  $S$  layers may exchange relative positions giving rise to  
58 polymorphism (Capitani, 2019).

59 On the other hand, bastnäsite and synchysite are the most important ore minerals for Ce, La, Nd, and Y.  
60 The demand for these REEs has spiked in recent years due to their increasing usage in numerous high-  
61 technology applications, including electronics and green technologies. For instance, Ce oxide ( $\text{Ce}_2\text{O}_3$ )  
62 is used in catalytic converters, La and Nd are used in the manufacturing of hybrid and electric motors  
63 and rotors of wind turbines, Nd compounds are used for the manufacturing of the most powerful  
64 permanent magnets occurring in microphones, speakers and hard disks, and synthetic Y garnet  
65 ( $\text{Y}_3\text{Al}_2\text{O}_{12}$ ) is used in filters for microwaves, acoustic transmitters and transducers, LEDs, lasers and  
66 even as gems (e.g., Goonan, 2011; Charalampides et al., 2015).

67 In nature, CRFC rarely occur as single crystals. Commonly, they form microscale syntactic  
68 (crystallographically oriented) intergrowths (Donnay & Donnay, 1953) of different  
69 polysomes/polytypes, often with stacking faults at the nanoscale. Due to this recurrent microstructure,  
70 definitive structural analyses by single-crystal X-ray diffraction (SCXRD) have been achieved  
71 relatively recently and only for some basic polysomes, namely bastnäsite-(Ce) (Ni et al., 1993),  
72 synchysite-(Ce) (Wang et al., 1994), and parisite-(Ce) (Ni et al., 2000). Apart from these fortunate  
73 cases, for most of the occurrences with intergrowths at the microscale, reliable structural analysis can  
74 only be performed via TEM.

75 Regarding SEM-EDS analysis, which is a relatively faster characterization technique compared to  
76 HRTEM, syntactic intergrowths can be revealed by the average atomic number ( $Z$ ) contrast in BSE  
77 images. However, no orientation relationships can be obtained, and submicroscopic intergrowths may  
78 be hidden and merged into a uniform-gray-tone microscopic band. Along the same lines, microprobe  
79 analysis, whose spatial resolution is on the order of a few micrometers, in the case of submicroscopic  
80 lamellae within the analyzed volume can only give an average composition that may accidentally  
81 match the composition of a potential polysome.

82 Raman spectroscopy is another relatively faster characterization technique that has been so far poorly  
83 exploited in these minerals. Ce-dominant CRFC has been investigated by Frost and Dickfos (2007),  
84 Guastoni et al. (2009), and recently by Zeug et al. (2021), sometimes providing contrasting results.  
85 Raman spectroscopy has a spatial resolution comparable to microprobe analysis, and according to Zeug  
86 et al. (2021), it can distinguish between some basic polysomes; however, the influence of  
87 submicroscopic lamellae on the Raman response is not clear. On the other hand, electron backscattered  
88 diffraction (EBSD) has, in principle, a spatial resolution that is much better (tens of nm) than that of the  
89 techniques mentioned above, and it may give orientation relationship information; however, its  
90 capability to distinguish between different CRFC has never been tested.

91 In this paper, we investigate well-characterized CRFC from Mount Malosa (Malawi) (Guastoni et al.,  
92 2009, 2010; Capitani, 2019) by combining Raman spectroscopy and EBSD—the latter applied, as far  
93 as we know, for the first time to CRFC—to evaluate the potentialities of these two methodologies in  
94 the characterization of this material at a microscopic scale and provide a “road map” for further focused  
95 investigation, aiming, for instance, to determine new polysomes with TEM.

96 Investigations at the microscopic and submicroscopic scale of CRFC (or any other ore mineral) may  
97 prove to be important for the understanding of the ore mineral formation and possibly for the  
98 improvement of downstream processing and REE recovery.

## 99 100 **Samples and Methods**

### 101 102 **Samples and sample preparation**

103  
104 The samples studied in this work come from Mount Malosa, Malawi, and were previously described by  
105 Guastoni et al. (2009, 2010) and Capitani (2019). All CRFC samples display a yellowish-reddish color  
106 and are associated with aegirine (Fig. 2). Before instrumental investigations, all samples were  
107 embedded in epoxy resin and then cut in parallel either to main or generic orientations, first roughly

108 determined by visual inspection of the crystal habit, then following the EBSD results (see ahead): i)  
109 sample 9c1 sectioned parallel to the x-y (z-axis perpendicular to the section); ii) samples 3 and 9c2  
110 sectioned parallel to the y-z plane (z-axis in the plane of the section); iii) sample 9b and 6 sectioned  
111 parallel to a generic orientation (z-axis at a high angle and a low angle to the section, respectively). All  
112 samples were mechanically polished with a rotational system using alumina with a nominal grain  
113 diameter of 0.3  $\mu\text{m}$  as the last polishing step. Those designated for EBSD investigations were further  
114 etch-polished with a Saphir Vibro vibratory polishing device using 0.06  $\mu\text{m}$  colloidal silica. Electron  
115 transparent TEM mounts were prepared from sample 9c2, i.e., the one with the optimal orientation to  
116 study syntactic intergrowths, which alternate along the c-axis (Donnay & Donnay, 1953). After SEM  
117 analyses, a double-polished 30- $\mu\text{m}$  thick section stuck with Attack® to a glass slide was obtained from  
118 the same samples. Copper rings, 3 mm in diameter, were stuck with Araldite® on the selected sample  
119 areas, removed from the glass along with the attached minerals by acetone dissolution of the Attack,  
120 and ion-milled down to electron transparency by a Gatan PIPS II Cool instrument. All samples were  
121 carbon coated with a 20 nm C-film before SEM observations and with a 5 nm C-film before EBSD and  
122 TEM investigations. The C-film was removed before Raman spectroscopy to avoid fluorescence.

## 123 124 **Instrumental analyses**

125  
126 SEM imaging and analysis were performed at the Platform of Microscopy of the University of Milano-  
127 Bicocca (PMiB) with a field emission gun (FEG) SEM Zeiss Gemini 500, operating at 20 keV and  
128 equipped with a Bruker XFlash EDS. The standardless method and ZAF correction were used for semi-  
129 qualitative analysis. EBSD investigations were performed with a high-resolution Bruker eFlash  
130 detector mounted on the same FEG-SEM Zeiss Gemini 500.

131 The Raman spectroscopy analyses were performed at the Department of Earth Science “Ardito Desio”  
132 of the University of Milan. The analyses were obtained at room temperature using a LabRAM HR

133 Evolution spectrometer. The system is equipped with an Olympus BX series optical microscope, a  
134 diffraction grating of 1800 grooves per millimeter, and a Si-based Peltier-cooled CCD detector. Spectra  
135 were excited with the 633 nm emission of a He-Ne laser and were obtained using a 100x objective with  
136 an acquisition time of 3x30 s. All the spectra were collected close to SEM analysis spots. Fitting of  
137 Raman spectra was done after background correction assuming Lorentzian band shapes. The system  
138 was calibrated using the  $520.7 \text{ cm}^{-1}$  line of a silicon wafer. A  $\frac{1}{2} \lambda$  wave plate was used to polarize the  
139 light. Spectra were obtained with two different polarization directions of the incident electron field  
140 vector ( $\mathbf{E}$ ):  $\mathbf{E} \perp z$  and  $\mathbf{E} // z$  in samples 3 and 9c2;  $\mathbf{E} // y$  and  $\mathbf{E} \approx // x$  in sample 9c1.

141 TEM observations were performed at the PMiB with a Jeol JEM2100Plus, supplied with a  $\text{LaB}_6$  source  
142 and operating at 200kV. The instrument is equipped with an Oxford EDS system and a Gatan Rio  
143 CMOS camera. The Digital Micrograph® software (Gatan) was used for image acquisition and  
144 processing. The HRTEM filter developed by D.R.G Mitchell (2007) was employed to reduce the  
145 inelastic scattering in HR images. EDS analyses were collected and quantified with Aztec (Oxford)  
146 software using the standardless method and correction for absorption. The method developed by Van  
147 Cappellen and Doukhan (1994) was used to estimate the thickness of the TEM mount at the point  
148 analysis spots. The estimated beam diameter at the sample surface was 3.5 nm.

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## Results

151

### Microstructure and composition (SEM-EDS results)

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153  
154 Syntactic intergrowths in CRFC were first studied by SEM-EDS for a first glimpse of the  
155 microstructure and the chemical variability at a microscopic scale. As expected, BSE images  
156 demonstrate the parallel banding typical of CRFC syntactic intergrowths (Fig. 3). EDS spot analyses  
157 were acquired within bands showing homogeneous gray tones. These analyses align with those

158 acquired by wave dispersive X-ray spectroscopy (WDS) in a previous study of different crystals from  
159 the same samples (Guastoni et al. 2009). Therefore, this indicates that EDS is sufficiently accurate for  
160 the identification of microscopic CRFC polysomes; at the same time, it is much faster than WDS  
161 (Table 1). The EDS analyses display an inverse correlation of the Ca/(Ca+REE) ratio with the BSE  
162 intensity (brightness) of the bands. All the measured phases are Ce-dominant and contain La and Nd as  
163 other major REEs, whereas Sm, Y, Pr, and Gd may be present as minor components. An exception is  
164 represented by synchysite, where Y is more abundant than La and Nd. Fluorine is underestimated in  
165 SEM-EDS analyses due to its tendency to diffuse under the influence of a highly focused electron  
166 beam. Overall, the chemical compositions of lamellae span from bastnäsite to synchysite depending on  
167 the sample and align almost continuously between röntgenite, parisite, and the  $B_2S$  polysome (Fig. 3).  
168 In particular, other than compositions close to bastnäsite, parisite, röntgenite and synchysite [ideal  
169 Ca/(Ca+REE) ratio of 0, 0.33, 0.40 and 0.50, respectively], lamellae with Ca/(Ca+REE) of 0.23 and  
170 0.27, which are close to the  $B_2S$  polysome (0.25), and lamellae with a Ca/(Ca+REE) of 0.36 (Table 1S,  
171 in the supplementary materials) have been often encountered. Moreover, a few analyses with a  
172 Ca/(Ca+REE) ratio of 0.20, theoretically corresponding to polysomes  $B_3S$ , 0.14 ( $B_5S$ ), and 0.29  
173 (intermediate between parisite and  $B_2S$ ), have also been collected. At this level, it is unknown whether  
174 or not the determined compositions correspond to ordered polysomes or arise from disordered  
175 intergrowths at the nanoscale.

176  
177 **Lamellae orientation (EBSD results)**  
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179 EBSD analyses were undertaken to i) establish the initial orientation of the samples, ii) align the  
180 sample for oriented cuts in view of both Raman and TEM investigations, and iii) test the method's  
181 capability to discriminate different polysomes. Unsurprisingly, the EBSD results allowed us to  
182 establish the sample orientation, set up the oriented cuts, and ascertain the effective syntactic

183 relationship among all the detected CRFC phases; however, it failed to distinguish between different  
184 polysomes (Fig. 4). This drawback can be easily understood if one considers that electron diffraction  
185 (similar to X-ray diffraction) is dominated by heavy atoms and that CRFC of the *BS* series, although  
186 monoclinic, have a hexagonal stacking of heavy atoms (Ca and REE) that is identical to that in  
187 bastnasite, the only effective hexagonal term (Ni et al., 1993, 2000; Wang, 1994). Different polysomes  
188 can be actually distinguished through EBSD maps if EDS chemical information is acquired at the same  
189 time and if the map is elaborated with both contributions (50% EDS and 50% EBSD). In this case, the  
190 syntactic intergrowths are correctly indexed, but the method loses interest since EDS has a spatial  
191 resolution (few  $\mu\text{m}$ ) two orders of magnitude worse than EBSD (tens of nm).

## 192 **Raman spectroscopy**

193 Raman spectroscopy analyses were taken on lamellae previously characterized by SEM-EDS in an  
194 attempt to correlate the Raman signal with the chemical composition, aiming to distinguish between  
195 different polysomes. Peak assignment was done according to White (1974), Buzgar and Apopei (2009),  
196 and Zeug et al. (2021). In particular, the following internal vibrational modes of the carbonate group  
197 were identified: i) symmetric stretching [ $\nu_1(\text{CO}_3)$ ]; ii) out-of-plane bending [ $\nu_2(\text{CO}_3)$ ]; iii)  
198 antisymmetric stretching [ $\nu_3(\text{CO}_3)$ ]; iv) in-plane bending [ $\nu_4(\text{CO}_3)$ ] (Table 2 and Fig. 5).

201 Since the intensity of Raman peaks changes in relation to both crystal orientation and laser polarization,  
202 four different configurations whose details and related Porto's notations (Damen et al., 1966) are  
203 reported in Table 3 were investigated. The highest intensity of the  $\nu_1(\text{CO}_3)$  stretching vibration was  
204 obtained in sample 9c2 with the  $x \begin{pmatrix} z \\ y \end{pmatrix} \bar{x}$  configuration. Conversely, the lowest intensity was obtained  
205 with the  $z \begin{pmatrix} y \\ x \end{pmatrix} \bar{z}$  setting (Fig. 6). Orientation of the polysomes and laser polarization influence the  
206 intensity of the bands only and not their positions.



207 In agreement with Zeug et al. (2021), we found that the  $\nu_4(\text{CO}_3)$  in-plane bending is in the range of  
208  $665\text{--}754\text{ cm}^{-1}$  and seems separated into two sub-regions. The  $\nu_2(\text{CO}_3)$  out-of-plane bending is around  
209  $870\text{ cm}^{-1}$ . The  $\nu_3(\text{CO}_3)$  antisymmetric stretching is around  $1440\text{ cm}^{-1}$  and is orientation dependent. The  
210  $\nu_1(\text{CO}_3)$  symmetric stretching is around  $1100\text{ cm}^{-1}$  and is split into three bands in intermediate  
211 polysomes: at  $\sim 1081\text{ cm}^{-1}$ ,  $1091\text{--}1095\text{ cm}^{-1}$ , and  $\sim 1099\text{ cm}^{-1}$ , whose relative intensities vary with  
212 composition (i.e., Ca/REE ratio, s. also Fig. 15). End members behave somewhat differently: bastnäsite  
213 demonstrates only one intense peak at  $\sim 1095\text{ cm}^{-1}$  and synchysite two bands at  $1081\text{ cm}^{-1}$  and  $1099\text{ cm}^{-1}$   
214 (Table 2 and Fig. 7). As observed in Zeug et al. (2021), we also detected other bands at 598, 1564 and  
215  $1738\text{ cm}^{-1}$ , whose origin was not clarified (Fig. 5).

216

### 217 **Nanostructure (TEM results)**

218

219 The sample studied by SEM-EDS and Raman spectroscopy with a favorable orientation for the study of  
220 syntactic intergrowths (i.e., with the z-axis laying on the observation plane, sample 9c2) was prepared  
221 for TEM to elucidate the structural state associated with the encountered compositions. Ordered regions  
222 in sample 9c2 are rare and limited to less than  $1\text{ }\mu\text{m}$  along the stacking direction. Indeed, the most  
223 recurrent microstructure is given by a pervasive occurrence of stacking faults (e.g., Fig. 8a) affecting  
224 bastnäsite and parisite, these last by far the most abundant phases detected in the sample (Fig. 8 and 9).  
225 Limited regions of  $B_2S$  and  $B_5S$  showing consistent compositions (Table 2S in the supplementary  
226 material) were also detected (Fig. 10), in agreement with SEM-EDS. As for SEM-EDS, a few point  
227 analyses with a Ca/(Ca+REE) ratio of 0.29 – unrelated to any basic polysome – were measured by  
228 TEM-EDS; however, they could not be connected with the underlying structure/microstructure.  
229 Moreover, a long-range polytype with a repeat distance of  $\sim 32\text{ nm}$  was also observed (Fig. 11).  
230 Conversely, even if SEM-EDS and Raman spectroscopy suggest abundant röntgenite, clear evidence of  
231 this polysome has not been confirmed by TEM. Along the same lines, the abundant lamellae with a

232 Ca/(Ca+REE) ratio of 0.36 detected by SEM-EDS (Table 1S) were not detected by TEM. Finally, a  
233 new parisite-(Ce) polymorph was detected, distinguished from normal parisite (Ni et al., 2000) and the  
234 other polymorphs described by Capitani (2019) by twice the repeat distance along  $c^*$  (~56 vs. ~28 Å,  
235 respectively), suggesting a repetition of four basic *BS* modules (~14 Å) along the stacking direction  
236 (Fig. 12).

237 Even considering that using TEM, it is not possible to explore the whole area explored by SEM and  
238 that we could have missed some compositions, our findings seem to indicate that, at least in part, the  
239 intermediate compositions detected by SEM-EDS on apparently homogeneous lamellae may result  
240 from sub-microscopic compositional faults (polysomatic faults), not resolved in BSE images, matching  
241 only by accident the composition of a theoretical polysome.

242 Compositional faults in bastnäsite can only be Ca-rich through the local insertion of vaterite-like slabs,  
243 whereas in parisite, they can be either Ca-rich or Ca-poor, i.e., through a bastnäsite-like slabs insertion.  
244 As matter of fact, in the studied samples, compositional faults in parisite are mostly Ca-poor. These  
245 conclusions are supported by nanoscale EDS analysis and HRTEM imaging (s. also Capitani, 2019).  
246 The Ca/(Ca+REE) ratio that is slightly higher than 0 in bastnäsite and lower than 0.33 in parisite (Table  
247 2S) can be interpreted in this way.

248 Figure 13 reports bright field (BF) scanning transmission electron microscopy (STEM) images and  
249 related compositional line scans across a Ca-poor lamella hosted in parisite and Ca-rich lamellae in  
250 disordered bastnäsite. In both cases, a clear inverse correlation between Ca and Ce can be observed.

251 Figure 14 reports an HRTEM image of bastnäsite ( $c \sim 10$  Å) including some compositional faults,  
252 consistent with  $B_2S$  and  $B_3S$  single slabs, with a thickness of ~19 and ~24 Å, respectively. The random  
253 occurrence of Ca-bearing compositional faults may explain the minor Ca content sometimes detected in  
254 bastnäsite and may cause an accidental matching of SEM-EDS analyses with intermediate polysome  
255 compositions, causing misinterpretation.

256

257

## Discussion and Conclusions

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### Polysome detectability through Raman spectroscopy

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260  
261 According to Raman results, the identification of the basic CRFC is possible through the symmetric  
262  $\nu_1(\text{CO}_3)$  stretching vibration at  $\sim 1100 \text{ cm}^{-1}$  (Fig. 5). Bastnäsite and synchysite can be easily  
263 distinguished from other polysomes by the strong Raman band at  $1095 \text{ cm}^{-1}$  present in the former and  
264 the two bands at  $1081 \text{ cm}^{-1}$  and  $1099 \text{ cm}^{-1}$  present in the latter. Parisite,  $B_2S$ , and röntgenite all have  
265 three different bands at  $\sim 1081 \text{ cm}^{-1}$ ,  $\sim 1091\text{--}1095 \text{ cm}^{-1}$  (hereafter  $1091 \text{ cm}^{-1}$ ), and  $\sim 1099 \text{ cm}^{-1}$ , which  
266 only differ in intensity. According to Zeug et al. (2021), the identification of these intermediate  
267 polysomes is possible through the  $1091 \text{ cm}^{-1}/1081 \text{ cm}^{-1}$  intensity ratio. We found this ratio to decrease  
268 linearly with the  $\text{Ca}/(\text{Ca}+\text{REE})$  ratio for samples with  $\mathbf{E} // z$  and with a parabolic shape for samples  
269 with  $\mathbf{E} \perp z$  (Fig. 15).

270 The number and position of the symmetric  $\nu_1(\text{CO}_3)$  stretching vibration Raman bands depend on the  
271 valence and ionic radius (Adler & Kerr, 1963) of the neighboring  $\text{CO}_3^{2-}$  groups. Actually, two different  
272 types of  $\text{CO}_3^{2-}$ -layers are present in the CRFC structure: those in contact on both sides with CeF-layers  
273 (or *e*-layers) and those in contact with one CeF-layer on one side and one Ca-layer (or *g*-layers) on the  
274 other side (Donnay & Donnay, 1953). Only (symmetric) *e*-layers are present in bastnäsite (Yang et al.,  
275 2008), resulting in one strong band at  $1095 \text{ cm}^{-1}$ , whereas only (asymmetric) *g*-layers are present in  
276 synchysite (Wang et al., 1994), resulting in two different bands at  $1081$  and  $1099 \text{ cm}^{-1}$ . Both *e*-layers  
277 and *g*-layers are present in intermediate polysomes such as parisite,  $B_2S$ , and röntgenite (Ni et al., 1993,  
278 2000), leading to a tripartition of the symmetric  $\nu_1(\text{CO}_3)$  stretching vibration.

279 The impression gathered after TEM-EDS is that ordered regions in CRFC syntactic intergrowths from  
280 Malawi are smaller than expected according to SEM-EDS imaging. Consequently, compositional faults

281 at the nanoscale in bastnäsite and parisite—not resolved in SEM-BSE images—may affect the  
282 Ca/(Ca+REE) ratio, which can match the composition of a real polysome only by accident. In light of  
283 these results, Raman spectroscopy, whose spatial resolution is  $\sim 1 \mu\text{m}$ , needs to be critically re-  
284 examined. We believe that the Raman signal is sensitive to different abundances of *e*- and *g*-layers in  
285 the structure, whose proportions vary with composition and thus also with the polysome, but not to the  
286 order of the layers within the analyzed volume. Therefore, whereas Raman spectroscopy could be a  
287 valid and faster method to probe the Ca/(Ca+REE) content of fluorcarbonates, it does not appear to be  
288 possible to distinguish between ordered and disordered intergrowths with similar compositions.

### 289 290 **Polysome detectability through electron-beam-related techniques**

291  
292 Although the nanoscale disorder can be overlooked, SEM-EDS is the fastest technique for polysome  
293 identification. TEM-EDS remains the ultimate technique for polysome identification; however, it is  
294 time-consuming and provides only local information. Since most high-resolution TEMs have a limited  
295 tilt range, the sample needs to be pre-oriented before preparation; in this case, EBSD can be very  
296 useful. In this regard, EBSD can easily distinguish CRFC among other phases and correctly provides  
297 their orientation relationship; however, under routine application, it fails to distinguish between  
298 different polysomes.

299 At the TEM scale, other than basic CRFC, intermediate polytypes that have not yet been fully  
300 described in the literature have been identified. Among these are a  $B_2S$  polysome, a 32 nm long-range  
301 polytype, and a new parisite polymorph with a double *c* parameter compared to normal parisite.  
302 Further investigations are required for a full characterization of all these new structures.

### 303 304 **Implications for REE Ore Formation**

305  
306 In principle, under equilibrium crystallization conditions, every single fluid composition falling within

307 the bastnäsite-synchysite series can be accommodated by a proper proportion of *B* and *S* layers;  
308 therefore, this situation opposes the simultaneous crystallization of two similar phases of different  
309 compositions, as in the well-known case of the alkali feldspar system at high pressure (e.g., Winter,  
310 2001) when the fluid composition falls within the miscibility gap, promoting the simultaneous  
311 crystallization of a Na-rich and a K-rich feldspar upon cooling.

312 The inability of the system  $\text{CeFCO}_3\text{--CaCO}_3$  to form solid solutions was considered a factor explaining  
313 the presence of syntactic intergrowths in CRFC (Donnay & Donnay 1953). One crystal precipitates  
314 until the conditions in the solution have changed sufficiently for the next compound to separate out,  
315 which then crystallizes on the original crystal. Since the two species alternate, periodic and  
316 discontinuous changes in the conditions of the system must be postulated. Variations in the  $a\text{Ca}^{2+}$ ,  
317  $a\text{REE}^{3+}$ ,  $a(\text{CO}_3)^{2-}$ ,  $a\text{F}^-$ , and *T* of the solution may well be the controlling factors during crystal growth  
318 (Gysi & Williams-Jones, 2015). Therefore, it appears that syntactic intergrowths are the equivalent of  
319 zoning in crystal-chemical systems that cannot form solid solutions.

320 Bastnäsite-(Ce) and synchysite-(Ce) both occur at Mt. Malosa (Malawi) but in separate samples, i.e.,  
321 they are never observed in contact. Moreover, the chemical composition is remarkably different in  
322 terms of REE partitioning, i.e., synchysite is richer in Y (and poorer in Ce, La, and Nd) than bastnäsite  
323 and all the other intermediate terms (Table 1 and 1S). This could indicate that synchysite formed under  
324 different time-space physicochemical conditions than the other polysomes.

325 The most recurrent microstructure in Mt. Malosa CRFC is represented by rhythmic parisite-bastnäsite  
326 intergrowths with a number of more or less ordered intermediate polysomes. This microstructure is  
327 similar to that of the Olympic Dam Australia deposit (Ciobanu et al., 2022); however, a clear trend of  
328 compositions varying gradually from a basic polysome to the next through disordered domains has not  
329 been observed at Mt. Malosa. The observed microstructure suggests a primary growth mechanism in  
330 which fluorcarbonates crystallize from a fluid close to thermodynamic equilibrium whose conditions

331 quickly and repeatedly crossed the parisite–bastnäsite stability boundary, rather than a stepwise  
332 approach toward thermodynamic equilibrium.

333 According to Secco et al. (2007), the crystallization of CRFC at Mount Malosa occurred at ~1 kbar and  
334 300–400 °C. These data are fully consistent with the mineral–fluid stability diagrams for the Ca–REE–  
335 C–O–H–F system of Gysi and Williams-Jones (2015), from which it appears how the boundary  
336 between parisite–bastnäsite, which has a negative slope on the  $\log aF^-$  vs.  $\log a(CO_3)^{2-}$  diagram, can be  
337 easily crossed after small variations of either  $a(CO_3)^{2-}$  or  $aF^-$ , thus representing the most probable  
338 reasons for the departure from equilibrium conditions. It should be noted, however, that an increase of  
339  $a(CO_3)^{2-}$  in the fluid (or  $aF^-$ , or both), for instance, would cause the crystallization of parisite at the  
340 expense of bastnäsite (Fig. 16). The protracting of this situation, however, would cause an increase of  
341  $aREE^{3+}$  in the remaining fluid, therefore realizing the conditions for the crystallization of bastnäsite (or  
342 some other polysomes poorer in Ca than parisite). In other words, the crystallization itself may induce  
343 rhythmic changes in the fluid composition at the crystallization front leading to syntactic intergrowths,  
344 which therefore appear as a rather unavoidable fact in Ca-REE fluorcarbonates.

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447 positions where the 3 mm copper rings were placed to extract TEM samples.

448

449 Figure 3. Left: SEM-BSE image of sample 9c2 showing the syntactic intergrowths of different CRFC  
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451 dark gray corresponds to röntgenite, light gray to bastnäsite and intermediate gray tones to parisite and

452 other intermediate polysomes (black areas are voids). Right: Ca/(Ca+REE) vs. REE/(Ca+REE) plot of  
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474 Ca/(Ca+REE) ratios of 0.23 and 0.27, respectively, close to the  $B_2S$  polysome (0.25). Note the strong  
475 orientation dependence of the intensity:  $\nu_1(\text{CO}_3)$  is maximal when the laser is polarized parallel to the

476 z-axis ( $x \begin{pmatrix} z \\ y \end{pmatrix} \bar{x}$ , solid line) and minimal when the laser is polarized perpendicular to the z-axis  
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478

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490

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494 reflections showing a periodicity of ~53 Å (c), whose intensity is further modulated at ~5 Å (brace),  
495 suggest a main building block of ~53 Å based on single *B*-layers, which we may tentatively indicate as  
496 the thick, darker lamellae in (a) (arrows).

497

498 Figure 12. SAED patterns of parisite-(Ce) structures along  $[1\bar{1}0]$ : a) Ni et al. (2000); b) Capitani (2019)

499 (both are simulations obtained with the CrystalMaker® X software); c) experimental pattern of the new  
500 parisite polymorph detected in this study; inset: zooming of the 11*l* row to highlight the ~56 Å  
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502 new polymorph with a double *c* parameter and a stacking of four *BS* single units (~14 Å).

503

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507

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511 slabs with thickness along *c* of ~19 and ~24 Å, respectively. Other than affecting the slab thickness  
512 (approximate values in Å on the right), the CF causes a shift on the (001) plane (*s* = straight, *l* = left, *r*  
513 = right shift).

514

515 Figure 15. 1091 cm<sup>-1</sup>/1081 cm<sup>-1</sup> intensity ratio for röntgenite (Roe), parisite (Par) and *B*<sub>2</sub>*S* for **E // z**(a)  
516 and **E ⊥ z** (b).

517

518 Figure 16. Calculated mineral–fluid equilibria at 300 and 1 kbar for the stability of bastnäsite-(Ce) and  
519 parisite-(Ce) as a function of *a*F<sup>-</sup> and *a*CO<sub>3</sub><sup>2-</sup> (Gysi and Williams-Jones, 2015).

Table 1.

<b>Mineral phase</b>	<b>EDS (this study)</b>	<b>WDS (Guastoni et al., 2009)</b>
Bastnäsite	$(\text{Ce}_{0.52}\text{La}_{0.27}\text{Nd}_{0.13}\text{Pr}_{0.04}\text{Sm}_{0.01}\text{Gd}_{0.01})_{0.98}$ $(\text{CO}_3)\text{F}_{0.63}$	$(\text{Ce}_{0.51}\text{La}_{0.32}\text{Nd}_{0.09}\text{Y}_{0.01}\text{Pr}_{0.04}\text{Sm}_{0.01})_{0.98}$ $(\text{CO}_3)(\text{F}_{0.95}\text{OH}_{0.07})_{1.02}$
Parisite	$\text{Ca}_{0.96}(\text{Ce}_{1.04}\text{La}_{0.56}\text{Nd}_{0.27}\text{Y}_{0.01}\text{Pr}_{0.09}\text{Sm}_{0.02}\text{Gd}_{0.01})_{2.00}$ $(\text{CO}_3)_3\text{F}_{1.33}$	$\text{Ca}_{0.92}(\text{Ce}_{1.03}\text{La}_{0.53}\text{Nd}_{0.27}\text{Y}_{0.04}\text{Pr}_{0.10}\text{Sm}_{0.04})_{2.01}$ $(\text{CO}_3)_3(\text{F}_{1.70}\text{OH}_{0.30})_2$
Synchysite	$\text{Ca}_{1.04}(\text{Ce}_{0.35}\text{La}_{0.17}\text{Nd}_{0.13}\text{Y}_{0.21}\text{Pr}_{0.03}\text{Sm}_{0.02}\text{Gd}_{0.03})_{0.94}$ $(\text{CO}_3)_2\text{F}_{0.97}$	$\text{Ca}_{0.99}(\text{Ce}_{0.42}\text{La}_{0.22}\text{Nd}_{0.13}\text{Y}_{0.19}\text{Pr}_{0.04}\text{Sm}_{0.03}\text{Th}_{0.01})_{1.04}$ $(\text{CO}_3)_2(\text{F}_{0.79}\text{OH}_{0.21})_1$

Table 2.

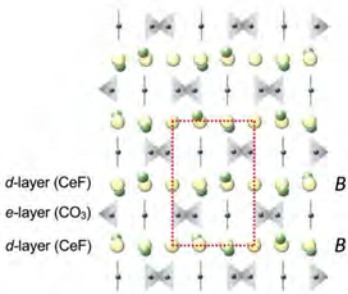
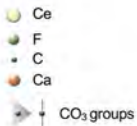
<b>Mode</b>	<b>Bastnäsite</b>	<b>Intermediate polysomes</b>			<b>Synchysite</b>	
$\nu_1$ - symmetric stretching	$\sim 1095 \text{ cm}^{-1}$	$\sim 1081 \text{ cm}^{-1}$	$\sim 1091\text{-}1095 \text{ cm}^{-1}$	$\sim 1099 \text{ cm}^{-1}$	$\sim 1081 \text{ cm}^{-1}$	$\sim 1099 \text{ cm}^{-1}$
$\nu_2$ - out-of-plane bending	$\sim 870 \text{ cm}^{-1}$					
$\nu_3$ - antisymmetric stretching	$\sim 1440 \text{ cm}^{-1}$					
$\nu_4$ - in-plane bending	$665\text{-}754 \text{ cm}^{-1}$					

Table 3

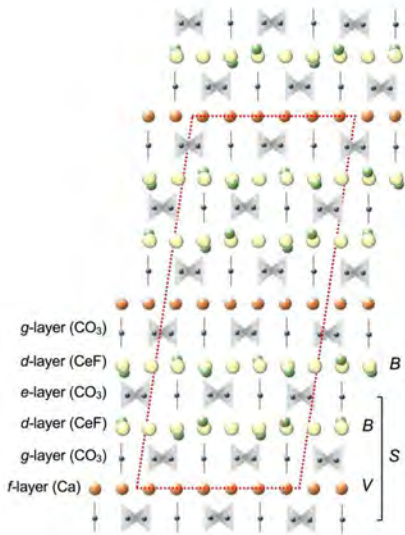
	Mineral section	Beam direction	E polarization	Porto's notation
1	$yz$	$x$	$z$	$x \begin{pmatrix} z \\ y \end{pmatrix} \bar{x}$
2	$yz$	$x$	$y$	$x \begin{pmatrix} y \\ z \end{pmatrix} \bar{x}$
3	$yx$	$z$	$y$	$z \begin{pmatrix} y \\ x \end{pmatrix} \bar{z}$
4	$yx$	$z$	$x$	$z \begin{pmatrix} x \\ y \end{pmatrix} \bar{z}$



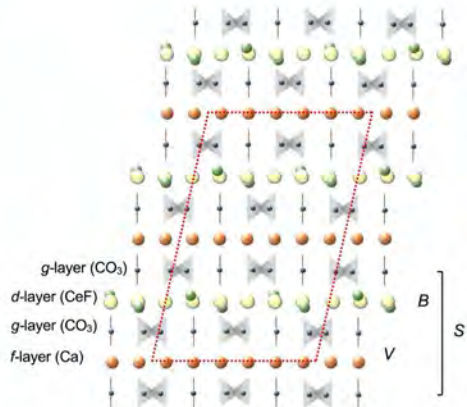
Figure 1



bastnäsite ( $B_2 - BB$ )



parisite ( $BS - VBBVBB$ )



synchysite ( $S - VB$ )

Figure 2

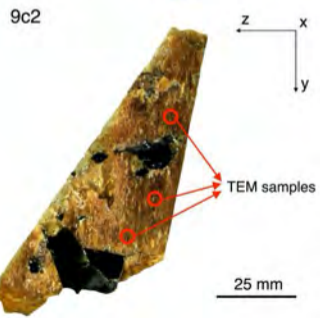
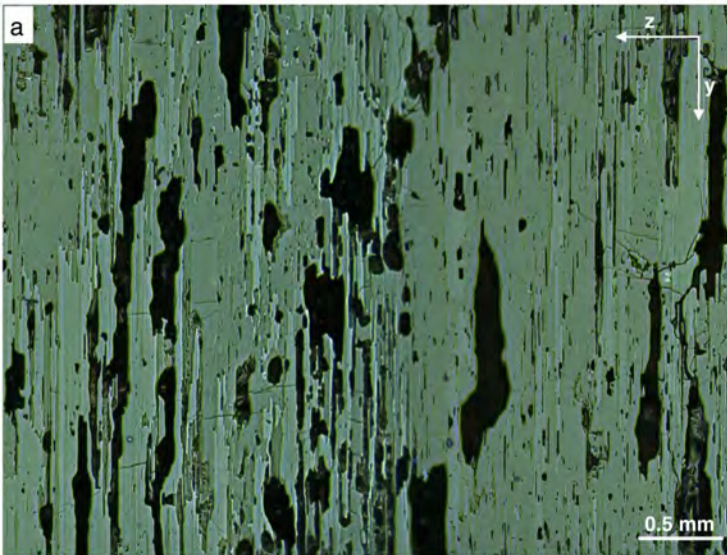


Figure 3

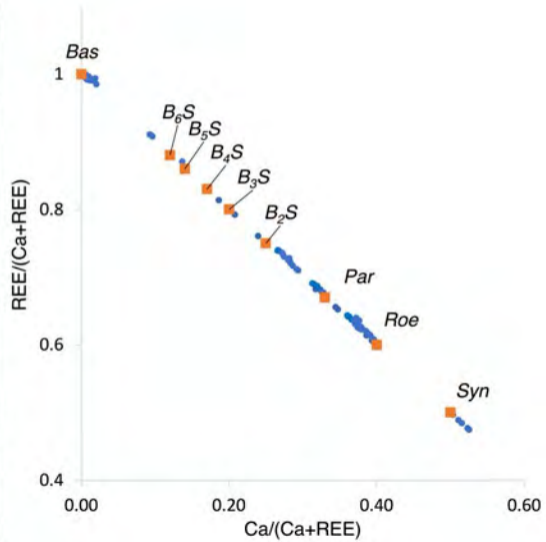
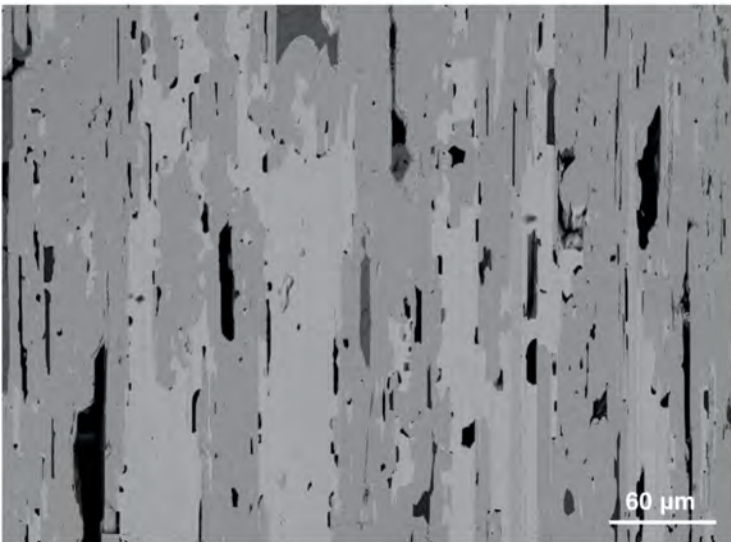
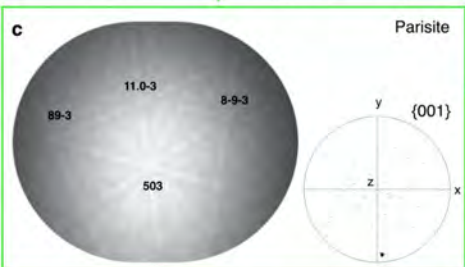
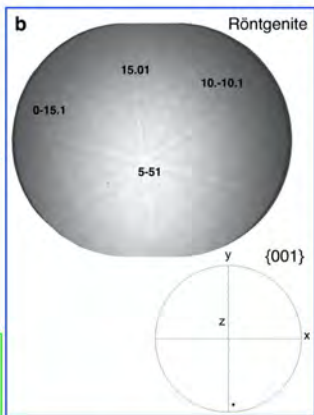
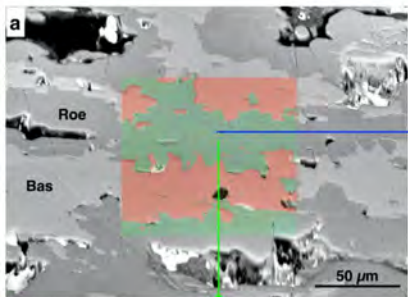


Figure 4



- Bastnäsite
- Parisite
- Röntgenite
- Synchronsite

Figure 5

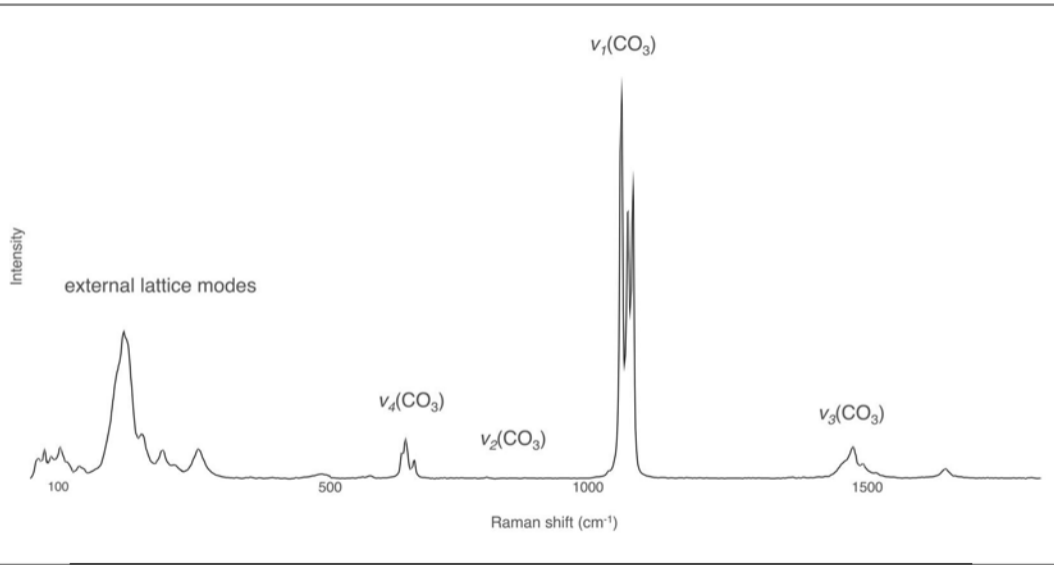


Figure 6

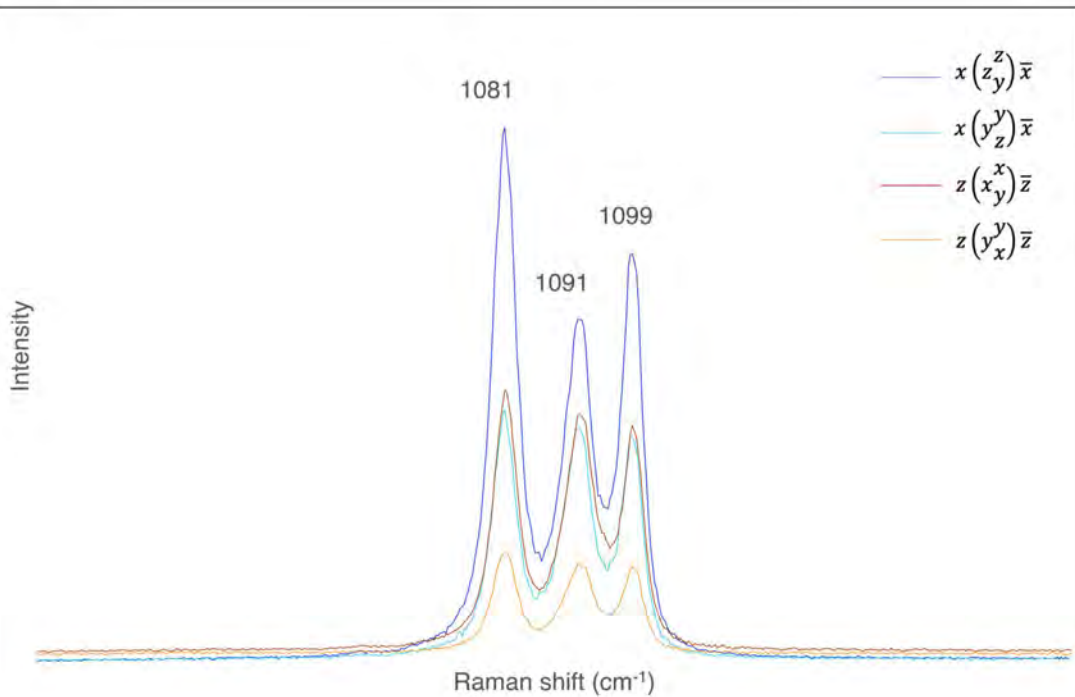


Figure 7

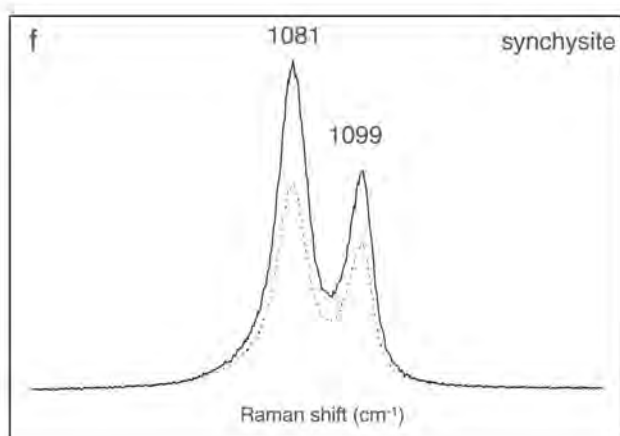
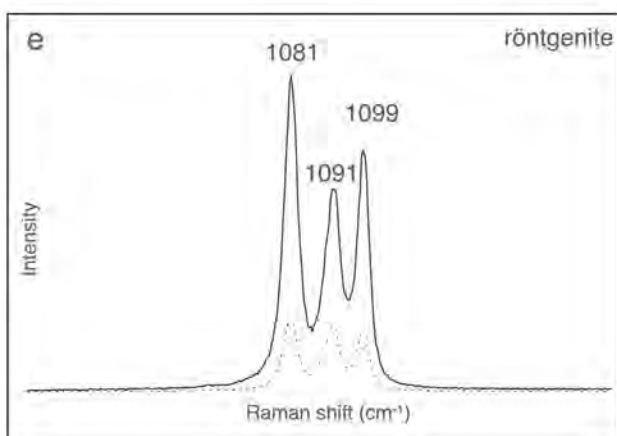
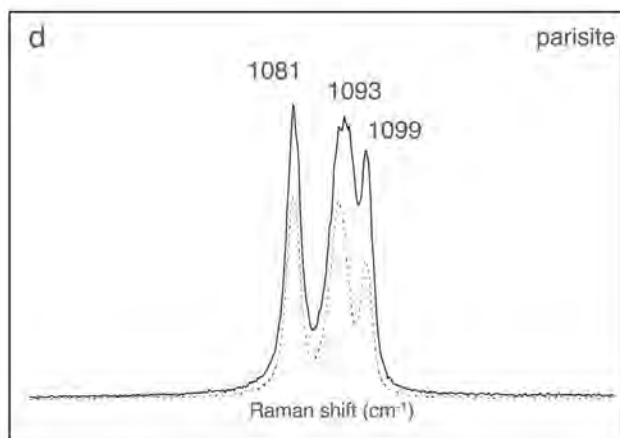
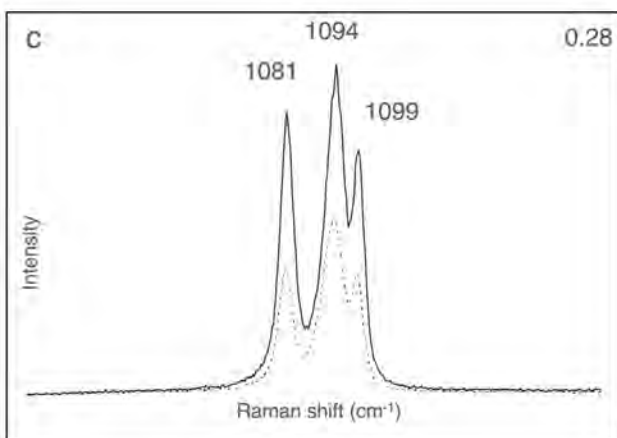
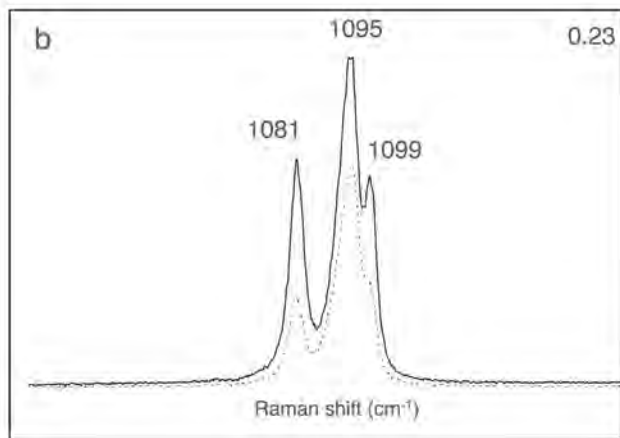
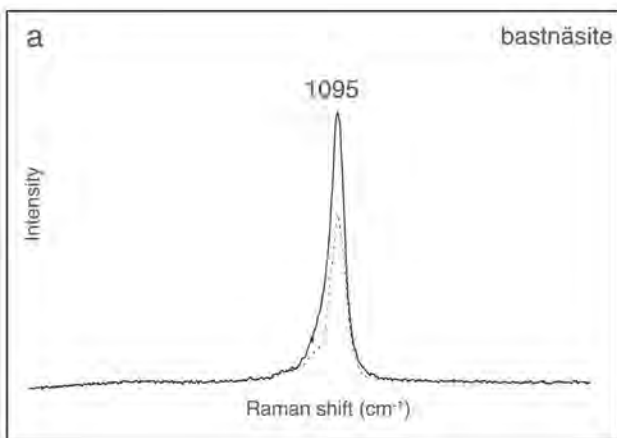


Figure 8

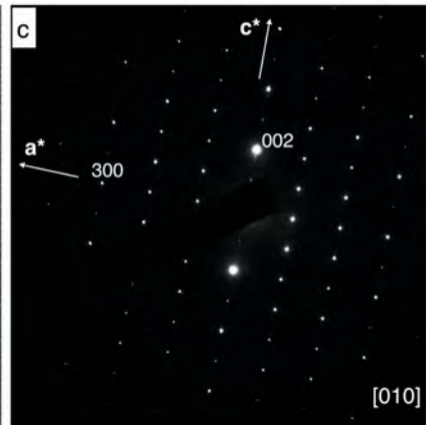
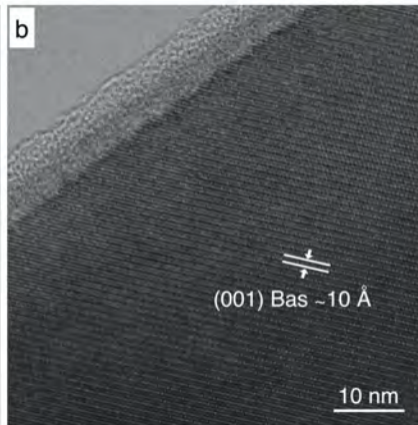
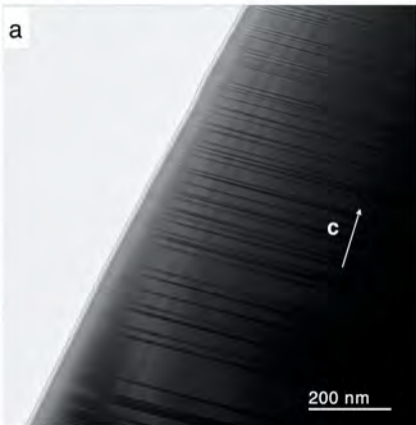




Figure 9

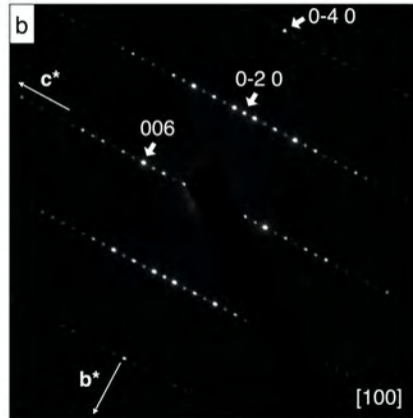
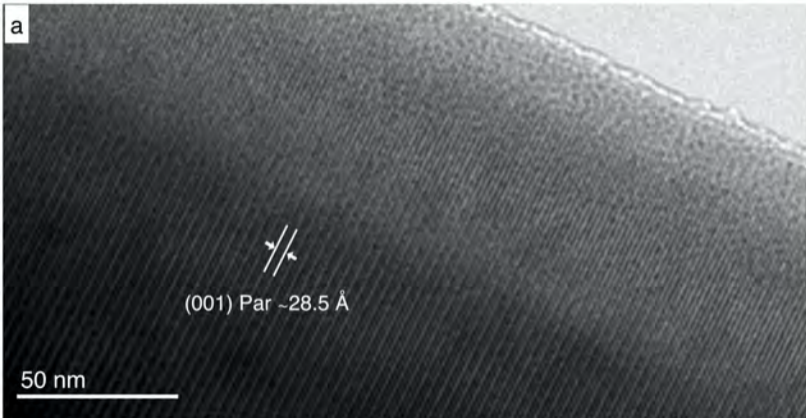


Figure 10

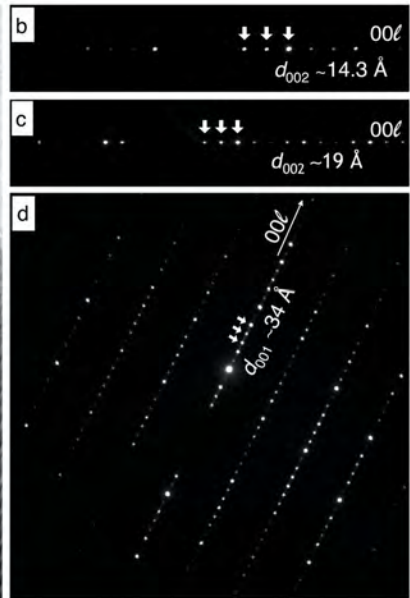
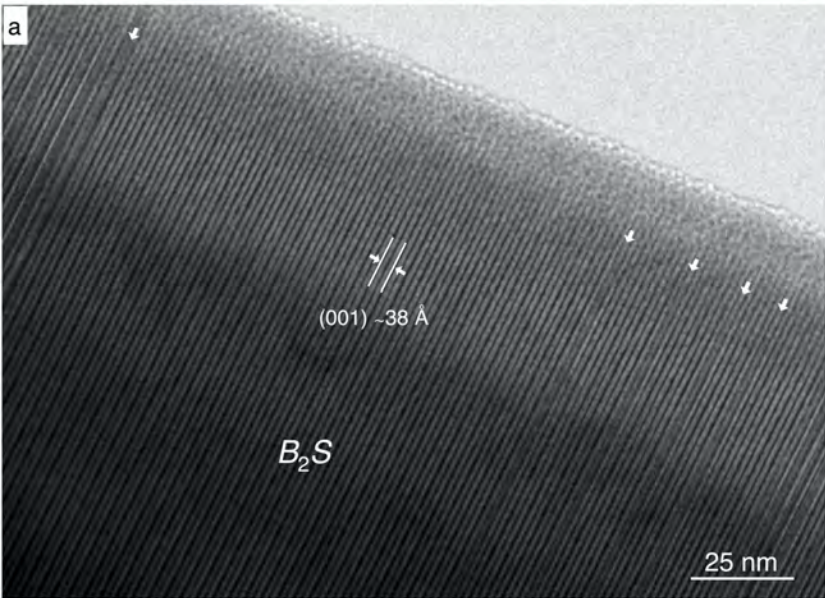


Figure 11

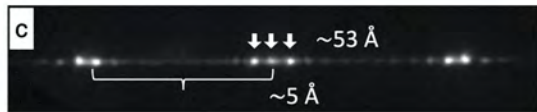
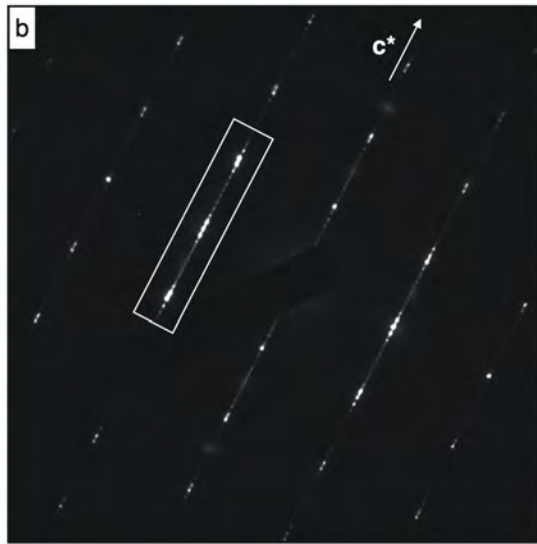
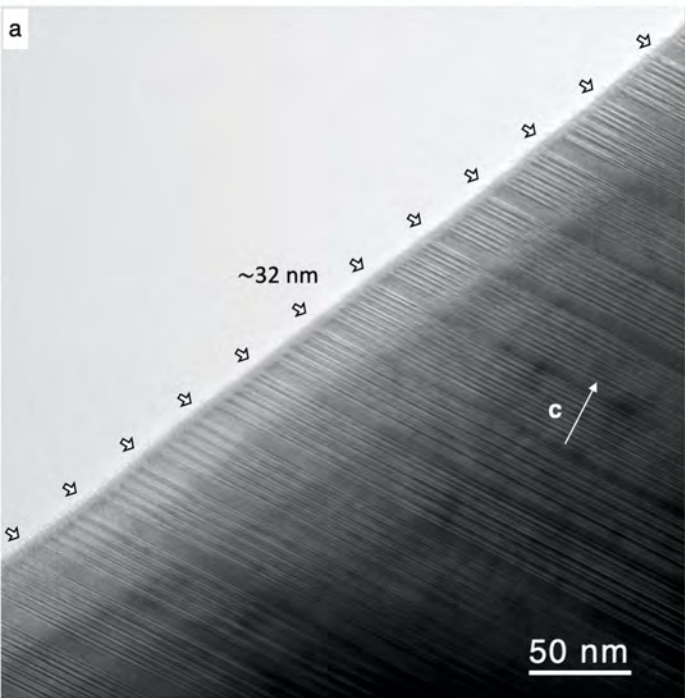


Figure 12

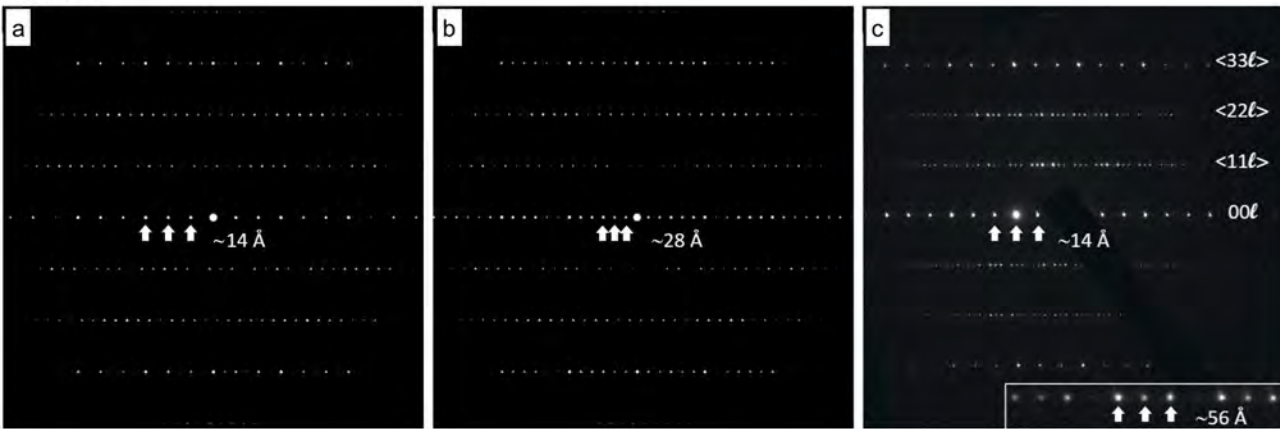


Figure 13

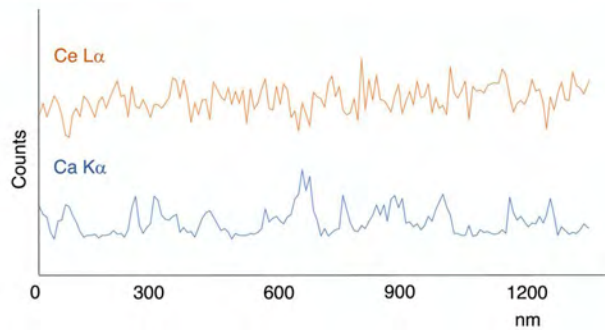
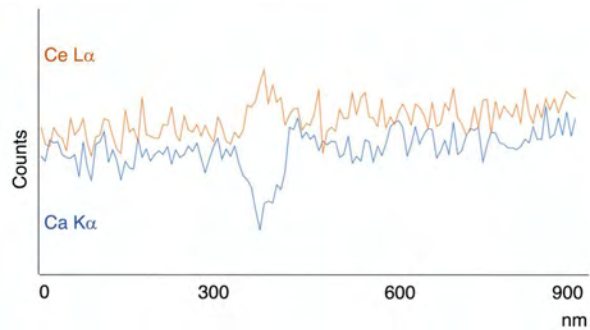
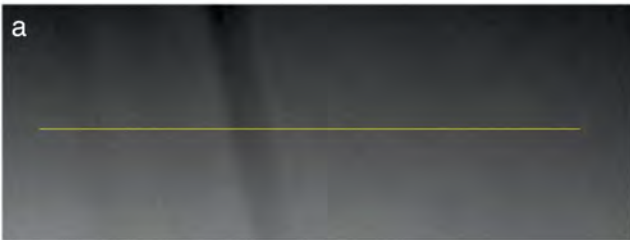




Figure 15

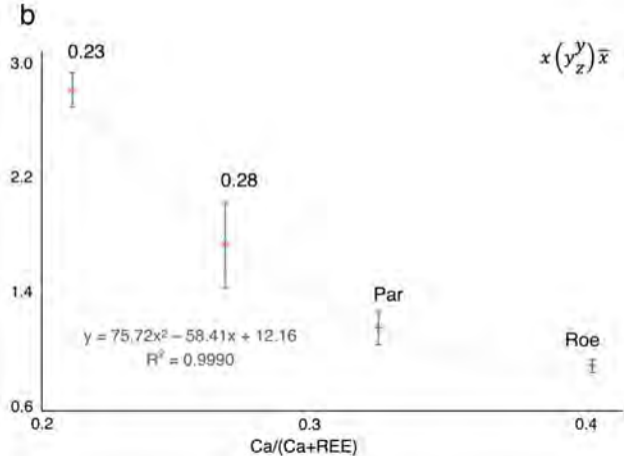
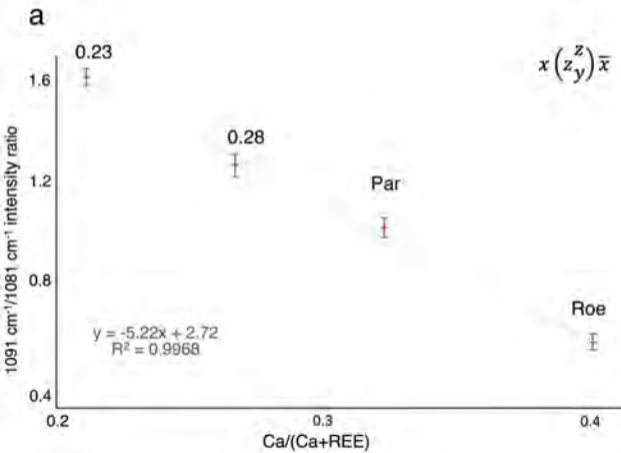


Figure 16

