

Ripplocations provide a new mechanism for the deformation of phyllosilicates in the lithosphere

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Abstract

Deformation in Earth's lithosphere is localised in narrow, high-strain zones. Phyllosilicates, strongly anisotropic layered minerals, are abundant in these rocks where they accommodate much of the strain and play a significant role in inhibiting or triggering earthquakes. Until now it was understood that phyllosilicates could deform only by dislocation glide along layers and could not accommodate large strains without cracking and dilation. Here we show that a new class of atomic-scale defects known as ripplocations explain the development of layer-normal strain without brittle damage. We use high-resolution transmission electron microscopy (TEM) to resolve nano-scale bending characteristic of ripplocations in the phyllosilicate mineral biotite. We demonstrate that conjugate delamination arrays are the result of elastic strain energy release due to the accumulation of layer-normal strain in ripplocations. This work provides the missing mechanism necessary to understand phyllosilicate deformation with important rheological implications for phyllosilicate bearing seismogenic faults and subduction zones.

Introduction

Layered phyllosilicate minerals (or sheet silicates) include clays, serpentine and micas and are important constituents of Earth's lithosphere, occurring in a range of rock types and tectonic settings. Laboratory studies show that interconnected layers of phyllosilicates are weak^{1,2}, while in the field these minerals are observed to be ubiquitous in plate bounding faults and shear zones^{3,4}. Due to their weakness, phyllosilicates are recognised to localise deformation and their rheology has a strong influence on the nucleation and propagation of earthquakes in seismogenic faults⁵⁻⁷ and on the dynamics of subduction zones and zones of shear in the viscous lower-crust^{3,8}. The mechanical response of phyllosilicates is due primarily to the strongly layered, and therefore anisotropic structure of their crystal lattice. This consists of layers of silicon-oxygen tetrahedra (T) and octahedrally coordinated cations (M) referred to as TMT layers (Fig. 1). In micas and most clays these layers are bound together by charge-balancing interlayer cations (such as K in biotite - Fig. 1a) and possess an overall negative charge. In serpentines and most chlorites tetrahedral-octahedral layers

34 are linked by Van der Waals and hydrogen bonding and their charge is neutral (Fig. 1b and c). The
35 layering described makes phyllosilicates particularly weak when sheared parallel to layers (the basal
36 or 'c' planes)⁹ and gives them a disproportionately large influence on the strength of the rocks in
37 which they occur^{3,10-14}.

38 Currently, viscous deformation in most minerals is understood to occur by the motion of defects
39 called dislocations through a process known as dislocation creep. This process is a combination of
40 dislocation glide, where the defects move along specific layers and directions in the crystal lattice
41 (known as slip systems) and dislocation climb, or cross-slip whereby dislocations are able to step
42 between planes in a crystal lattice to avoid obstacles such as impurities. The strongly layered
43 structure of phyllosilicates, however, limits dislocation glide to the basal plane with no facility for
44 dislocation motion on other planes⁹. As a result, dislocation creep is not a viable deformation
45 mechanism in phyllosilicates and the means by which strain is accommodated parallel to the c-axis
46 (perpendicular to the layering) remain ambiguous. In addition to this, the work of Noe and Veblen¹⁵
47 questions the viability of dislocation defects within the (001) biotite interlayer (the cleavage plane)
48 altogether, on the basis of energy considerations. These authors show that instead, basal
49 dislocations are found only within the (001) oxygen layer between the octahedral and tetrahedral
50 sheets, thus highlighting that it is a misconception to assume that cleavage planes are glide planes
51 for dislocations.

52 In layered minerals kinking is observed to be the dominant process of shortening parallel to basal
53 planes (Supplementary Figure 1 and 2). However, kink bands cannot be explained using glide of basal
54 dislocations alone. The most comprehensive models invoke the formation of complex arrays of
55 dislocation walls at kink band boundaries (KBBs) which impart curvature of the lattice over a finite
56 region^{16,17}. While these models determine basal slip as the primary mechanism for kinking, they
57 recognise that another mechanism is necessary to account for the c-axis parallel strain required to
58 form kink bands¹⁶ (KBs) (Supplementary Note 1). To date this c-axis strain has been explained

59 through the brittle damage mechanisms of dilation or cleavage when basal glide is inhibited^{17,18}. The
60 importance of phyllosilicates with regard to the strength and dynamics of the lithosphere demands a
61 proper understanding of their behaviour. Despite this, existing mechanisms fail to fully explain
62 commonly observed structures and processes associated with phyllosilicate deformation, suggesting
63 an important piece of the puzzle is missing.

64 Recently, a new fundamental defect type, named ripplocations, has been proposed which takes the
65 form of atomic-scale ripples in the basal layer¹⁹. They have so far been studied and modelled in
66 MoS₂¹⁹, Ti₃SiC₂^{20,21} and graphite^{21,22} but are theoretically applicable to phyllosilicates and other
67 layered solids²³. Ripplocations have a similar effect to dislocations in that they allow the motion of
68 one plane of atoms over another and may be more favourable in certain crystallographic
69 directions^{19,24}, but differ because they do not require the breaking and re-building of in-plane bonds
70 (Figs. 1d and e) and result in an expanded unit cell dimension normal to the weak layer. Another
71 fundamental difference is that ripplocations attract each other and may merge, in contrast to same-
72 sign dislocations, which repel and pile-up^{19,21}. Ripplocations on different layers are also capable of
73 aligning to form a through-going bend in the lattice, i.e. a kink band boundary (KBB)^{21,23}. Crucially,
74 ripplocations involve a ripple in the basal layers, giving them a component of c-axis parallel strain
75 that is absent in basal dislocations. They have been visualized in transmission electron microscope
76 (TEM) observations of experimentally deformed MoS₂¹⁹ and Ti₃SiC₂^{20,21}, while ripplocation behaviour
77 has been tested through numerical modelling^{19,21}. However, prior to this study, ripplocations have
78 not been reported in any naturally occurring mineral. The geological implications of the presence of
79 ripplocations in phyllosilicates are substantial.

80 They provide a previously unrecognised mechanism to account for the geometric problems
81 associated with deforming such anisotropic minerals. Phyllosilicates are increasingly seen as
82 fundamental players in the localisation of strain in the lithosphere, with a controlling effect on the
83 rheology of many crustal rocks^{3,7,8}. Our current understanding of the deformation of these important

84 minerals is hindered by the lack of a mechanism for geometrically necessary c-axis strain.
85 Ripplcation motion and their interactions may be that missing mechanism. In this study we use high
86 resolution TEM to examine the nano-scale structure of naturally deformed biotite mica from a
87 regional scale shear zone in the Ivrea-Verbano area of North-West Italy (Supplementary Note 2), and
88 show that ripplocations are essential to account for the lattice bending and basal delaminations we
89 observe.

90 **Results**

91 **TEM observations of ripplocations in biotite**

92 TEM samples were prepared using the focussed ion beam (FIB) in situ lift out technique²⁵ (see
93 methods). All biotite grains analysed contained regions of lattice expansion parallel to the basal
94 plane, these were up to 0.2 μm in length and visible as bright streaks in brightfield images (Fig. 2
95 Supplementary Figure 3). The width of biotite interlayers expands from 3 \AA in the undisturbed lattice
96 to up to 60 \AA in the regions of greatest lattice expansion. The TMT layers by contrast remain at 7 \AA
97 width but form ripples deflecting around regions of lattice expansion (Fig. 3). Expanded regions,
98 transitioning to delaminations in the brightest areas, are often asymmetrical, lozenge shaped and
99 occur in en-echelon stacks, which are aligned in two principal directions, 30°-60° apart and bisected
100 by the basal plane (Fig. 2a-d). This geometry produces an elongated diamond-shaped pattern across
101 entire biotite grains (Fig. 2a, Supplementary Figure 4). Expansion structures have the appearance of
102 waves (Fig. 3) and exist at multiple scales, from delamination of two TMT layers, along the interlayer,
103 visible at the micron-scale, to ripples that increase interlayer distances at the nano-scale (arrows in
104 Fig. 3a). Such large rotations of layers forming ripples are orders of magnitude greater than those
105 which could be produced by individual dislocations or complex arrays of dislocation walls, which can
106 result in bending of the (001) plane only within a broader finite region on the order of 1-2 μm ^{16,17}.
107 Bending of (001) within individual KBBs has been described on the scale of 40° in 0.2 μm ¹⁷ and was
108 interpreted to have been achieved by the rotation accumulated across numerous dislocation walls.

109 However, we observe rotation of very low numbers of (or in some cases individual) biotite TMT
110 layers on the order of 30° across just 3 or 4 nm (Fig. 3). According to the above studies, individual
111 dislocation walls are capable of rotating (001) by up to 2° , this would mean our observations require
112 around 15 individual dislocation walls within the space of 3 or 4 nm, a distance of less than 8 unit
113 cells in the a direction and around 4 unit cells in the b direction. This degree of rotation cannot be
114 physically accommodated by means of dislocation walls but can be explained by elastic bending of
115 basal layers as in existing models of ripplocations^{19,21}. Profiles drawn across expansion structures
116 concur that regular 10 Å spacings (the c-axis dimension of a biotite unit cell) increase, and that this
117 expansion occurs within the interlayer regions (Fig. 3a, b and c).

118 Most previous studies on interlayer delaminations in phyllosilicates did not utilise the FIB sample
119 preparation method but rather used ion milling systems which involve polishing until a perforation is
120 produced in the centre of the sample (See Methods). Therefore, for direct comparison with previous
121 work, and to eliminate the FIB method as a potential source of delamination structures, we also
122 prepared foils from 3 samples using an Ar precision ion polishing system (PIPS). At the ultra-thin
123 edges of the central perforation of PIPS specimens, voids could occasionally be seen to grow during
124 observation. They often became much wider than delaminations in FIB specimens or within the
125 thicker regions of PIPS samples and had lower aspect ratios. In contrast, thicker regions further away
126 from the perforation edge contained smaller expansion structures and delaminations closely
127 resembling those of the FIB specimens in size, aspect ratio and their arrangement in diamond-like
128 arrays (Supplementary Figure 5). To examine the relationship between these arrays and the degree
129 of strain a biotite grain has experienced, samples were prepared (using both FIB and PIPS methods)
130 from undeformed Westerly granite (WG) specimens to be compared directly to those from mylonitic
131 biotite described above and shown in Fig. 2 and Fig. 3 (Supplementary Table 1). Diamond-shaped
132 arrays of expansion structures and delaminations were present in all the biotite samples analysed
133 (Supplementary Figures 4-6) but were more clearly observed and more abundant in biotite from
134 naturally deformed mylonite samples. Evidence for ripplocations in nominally undeformed granitic

135 biotite is not entirely surprising and might be linked to two possible causes: 1) Rippllocations are a
136 form of crystal defect and might therefore develop during crystal growth, due for example to layer
137 mismatch, much like the way growth-related lattice mismatch forms dislocations²⁶; 2) An
138 undeformed granite, during intrusion through the Earth's crust, could develop local grain-scale
139 differential stresses due to crystal impingement during growth or differential thermal
140 expansion/contraction.

141 It must be noted that we did not observe kinking in biotite grains from Westerly granite, suggesting
142 that, while rippllocations are present in this biotite, no motion or interaction of rippllocations have
143 occurred. This observation may support the idea of rippllocations as growth defects (point 1). The
144 evident increased abundance of lattice expansion and delaminations in biotite from mylonites is
145 indicative of the intense deformation experienced by these rocks relative to granites.

146 The best explanation for the geometric arrangement of conjugate lattice expansion arrays we show
147 in this study is an overall expansion parallel to the c-axis (perpendicular to the basal plane) of the
148 biotite grain. The fact that the edges of these 'diamond' geometries are 30°-60° apart and are
149 bisected by the basal plane is not a coincidence, it is a clue to the stress field in which these
150 structures formed. The clear conceptual comparison is to conjugate normal faulting, which occurs
151 under extensional stresses, where σ_1 is vertical and σ_3 is horizontal and parallel to extension (Fig. 4a).
152 The acute angle between faults is commonly 60° but can vary and the effect on the rock mass is an
153 extension perpendicular to the plane that bisects this angle²⁷. In our diamond-shaped arrays, the
154 bisecting plane is the basal plane and the extension (and σ_3) would therefore be parallel to the c-axis
155 (Fig. 4b). These conjugate arrays are therefore likely to have formed during expansion parallel to the
156 c-axis (normal to the basal planes).

157 Since, in order to achieve electron transparency, TEM specimens are exceptionally thin (< 100 nm)
158 the possibility arises that the delaminations form to relieve stored c-axis strain once the grain
159 becomes completely unconfined during thinning²³. Basal dislocations are not a viable mechanism for

160 storing this strain energy as they possess no component of c-axis strain. We propose that this c-axis
161 strain could initially be stored in the form of bulk ripplocations, which originate from compression
162 parallel to the basal planes (as a component of the overall stress field) and their strain energy is
163 released to form conjugate arrays of expansion structures on thinning for TEM analysis. This
164 interpretation is supported by substantial flexing of the TEM films observed during FIB preparation
165 (Supplementary Figure 7), indicating elastic energy release.

166 Delaminations have been previously observed in TEM studies of deformed micas²⁸⁻³¹ and attributed
167 to electron beam damage²⁸. However, the extensive, ordered diamond-shaped arrays described here
168 have not been reported before. To assess whether the structures seen are caused, or extenuated by
169 beam damage, we analysed regions previously unexposed to the electron beam. The first evidence
170 that abundant delaminations are not beam damage artefacts is the fact that they are visible at the
171 outset of observations in the TEM and cover grains extensively and uniformly (Supplementary
172 Figure 8a). Beam effects were observed during our experiments, but took the form of progressive
173 mottling and amorphisation, eventually obscuring or destroying the lattice structure (Supplementary
174 Figure 8a, b). Beam damage effects are expected and observed to be localised to areas where the
175 beam has been held and should not occur uniformly across a whole grain.

176 The principal mechanism previously provided for the formation of basal delaminations is the
177 diffusion of alkali interlayer cations (Na in paragonite) due to beam exposure²⁸, with layer
178 separations forming to counteract the resulting volume decrease. This was supported by a rapid
179 decrease in the Na peak in sequential X-ray spectra of paragonite collected during progressive beam
180 exposure. Interactions between alkali elements and an electron beam result in a depletion of the
181 element's concentration at the sample surface and higher concentrations immediately below^{32,33}.
182 This must be accounted for during electron microprobe analysis of geological materials^{34,35}. In our
183 study, K is the interlayer cation in biotite, it is heavier than Na and, under the same beam conditions,
184 is also less mobile due to its lower diffusivity³². Despite this, the delamination arrays we document

185 are more extensive than those reported in paragonite (Fig. 1a of Ahn et al.²⁸) suggesting that
186 diffusion of alkali cations may not be the cause. Energy dispersive X-ray (EDX) spectroscopy of our
187 specimens in the TEM shows a decrease in K relative to Si over periods of up to 16 minutes, as could
188 be expected from cation diffusion³⁵, however the rate of decrease slows over time and stabilises
189 asymptotically around values of 0.33 K/Si (Supplementary Figures 8c, 9-13). This is in contrast to the
190 rapid and near-total loss of Na in the paragonite analysed by Ahn and co-workers²⁸. In addition, in
191 our observations K loss cannot be linked with a simultaneous nucleation or growth of delaminations
192 because these are present before any K loss occurs. While the beam damage mechanism²⁸ does not
193 therefore explain our structures, the delamination of basal planes must indeed have been driven by
194 some degree of c-axis parallel strain. As basal dislocations cannot impart c-axis strain we propose
195 ripplocations as the defects governing the observed (001) interlayer plane bending and associated
196 expansion.

197 **Discussion**

198 Our model for the formation of conjugate expansional arrays is in agreement with numerical
199 modelling of ripplocations in Ti_3SiC_2 and graphite²¹, which suggests ripplocations, when confined,
200 nucleate as multiple small ripples, but when unconfined, fewer, larger undulations are energetically
201 favoured. This is analogous to the production of delaminations in our samples when the
202 'confinement' is removed during thinning. In other words, the energy of the ripplocations is unable
203 to delaminate the layers around them until the confining energy is sufficiently reduced. Multiple
204 small ripplocations build up whilst the rock containing the biotite is experiencing differential stress
205 (Fig. 5a-b). If these stresses continue, or increase, then eventually the ripplocations gain enough
206 energy to overcome the constraining force (lithostatic pressure) and may migrate, interact or
207 accumulate across layers to form KBBs, hence the kinking of phyllosilicates in deformed rocks (Fig.
208 5c). If the ripplocations do not exceed the required energy to become mobile and form kinks, then
209 they continue to store that strain energy until the confining energy is reduced, for example during

210 TEM sample preparation. They then release c-axis strain energy with many small ripplocations on
211 the same plane merging to produce fewer, larger expansion structures or delamination features
212 which form in conjugate extensional arrays (Fig. 5d). Through this model, ripplocations provide the c-
213 axis driving force necessary to produce the basal delaminations observed in our samples and
214 reported in previous studies. The model also provides the c-axis strain necessary to explain the
215 pressure sensitivity measured in phyllosilicate-rich rocks (whereby their yield strength increases
216 proportionally to the applied lithostatic pressure) up to their dehydration temperature¹³. In
217 Phyllosilicates both viscous (dislocation glide) and brittle (cracks associated with kinking and dilation)
218 deformation mechanisms are seen to occur through a large range of conditions in the Earth (from
219 the Earth's surface to the middle and lower crust^{13,36-38}). In addition to this, while pressure sensitivity
220 might traditionally be associated with brittle deformation, it occurs in micas even when no obvious
221 microstructural evidence for classic brittle deformation can be observed¹³. Essentially, micas might
222 deform by a pressure sensitive but non-brittle mechanism at a range of conditions. This has to date
223 been a poorly understood phenomenon. Ripplocations resolve this conflict by providing a pressure
224 sensitive mechanism of deformation that does not result in fracturing or other characteristic brittle
225 microstructures (see also Supplementary Note 3).

226 In PIPS prepared samples, exposure to the beam did appear to initiate further distortion and growth
227 of voids in ultrathin regions close to the perforation edge. It is possible that the small amount of
228 energy imparted by the electron beam was enough to release further elastic strain from existing
229 ripplocations in these regions, especially where a free sample edge could be exploited. This still
230 requires the storage and release of some degree of c-axis strain which cannot be explained by basal
231 dislocations. The fact that these effects are only observed at ultrathin perforation edges and not in
232 thicker regions or in FIB specimens, could explain why previous studies, which utilise PIPS-style
233 methods, have reported large delaminations that grow under the beam²⁸ and have not reported
234 conjugate expansion structure arrays.

235 This work demonstrates the existence of ripplocations within the phyllosilicate mineral biotite and
236 highlights the fact that ripplocations may be a ubiquitous deformation mechanism in all types of
237 phyllosilicates as they are applicable to all layered solids²³. In particular, serpentine minerals and
238 chlorite may be even more susceptible to the nucleation and motion of ripplocations than micas.
239 Their structures do not incorporate interlayer cations and their interlayer bonding relies on weak
240 Van der Waals forces and hydrogen bonds³⁹ (Fig. 1b and c). In clay minerals the impact of
241 ripplocations may be enhanced by lower lithostatic pressures in the upper crust, where clays are
242 important constituents of fault gouges, as the size and mobility of ripplocations is likely a function of
243 confinement²¹.

244 The existence of ripplocations has fundamental implications for our understanding of phyllosilicate
245 deformation. Existing models of kink band formation struggle to explain c-axis parallel strain without
246 invoking brittle mechanisms. KBBs display a range of rotation angles and radii of curvature, and may
247 be partially elastic, as shown by reversible experimental hysteresis curves⁴⁰. Such 'incipient' kink
248 bands appear to be entirely reversible, up to a point where they are pinned and transition into
249 permanent kink bands. This process of recoverable plasticity is better explained by the formation
250 and destruction of ripplocations than by the motion of basal dislocations or dislocation walls. The
251 ability of ripplocations to store extensive elastic strain energy raises questions on how this strain
252 may be distributed to other phases or released under changing conditions in the Earth. It is worth
253 noting that, while studies of natural rocks represent a 'frozen in' microstructure in which the
254 contribution, or impact, of elastic strain is difficult to quantify post-mortem, elastic strains are likely
255 to play an important role in the development of locally high stresses with a significant effect on
256 recrystallisation and mineral reactions^{41,42}. It remains unclear whether ripplocations should be
257 defined as either brittle or viscous defects (see also Supplementary Note 3). They are defined as
258 waves in atomic layers¹⁸⁻²⁰ and can form at different scales, involving few to several interlayer bonds.
259 Ripplocations may stretch interlayer bonds (visco-elastic behaviour), but larger scale ripplocations
260 may cause these bonds to break (delaminations as lenticular micro-cracks). If lenticular micro-cracks

261 form, the motion of ripplocations might drive micro-crack migration in a continuous cycle of cracking
262 and healing, thus accommodating deformation via a combination of visco-elastic and transient
263 brittle processes. The behaviour we describe here would have considerable implications for fluid
264 transport in deep shear zones where ripplocations would provide the transient porosity waves
265 needed to move fluids through the crust. This idea is not dissimilar to the model proposed by
266 Morgan and Holtzman⁴³ where vug waves are defined as a combined deformation and fluid
267 migration mechanism whereby migration of fluid-filled cracks is driven by the release of elastic strain
268 energy. Fluids in faults, shear zones and subduction zones can weaken the mineral components of
269 these structures, increase the rate of diffusive mass transfer, or generate overpressures. While the
270 applicability of a vug wave-type model to ripplocations must be tested carefully with further work,
271 the implications for the strength of faults, shear zones and subduction zones are significant. As the
272 properties of ripplocation defects are clearly important for understanding the rheology of micas in
273 mylonites of the middle and lower crust, so they may also prove a critical consideration for the
274 frictional behaviour of phyllosilicates within seismogenic faults and subduction zones^{44,45}. Now that
275 ripplocations have been identified in natural phyllosilicates, these defects must be accounted for in
276 microphysical modelling of the mechanical properties of phyllosilicate-rich faults, shear zones and
277 subduction zones as they are likely to have a crucial influence on the rheology of these tectonic
278 lineaments and on the nucleation and propagation of large magnitude earthquakes^{5,6,44-46}.

279 Methods

280 **Preparation of TEM samples**

281 Electron transparent TEM specimens were produced by both focused ion beam (FIB) and Ar ion
282 milling in order to identify and compare any effects of sample preparation procedure on the
283 distribution and morphology of delaminations. This comparison is important as FIB is a popular and
284 effective method of TEM sample preparation however the majority of previous studies on deformed
285 phyllosilicates utilised ion milling techniques. The FIB specimens were produced using the lift out

286 method using a FEI Helios Nanolab™ 600 I DualBeam™ Ga focused ion beam (FIB) scanning electron
287 microscope (SEM). Target biotite grains were identified in standard petrographic thin sections of
288 mylonitic samples using optical and scanning electron microscopy. Site specific specimens were
289 prepared using the FIB, with sections cut perpendicular to cleavage via the in-situ trenching and lift
290 out technique²⁵ (Supplementary Figure 7a). Once prepared, thin lamellae received a 5.00 kV low
291 energy surface wipe, to reduce the thickness of amorphized surface damaged layers. During this
292 thinning process, all samples were observed to bend and flex extensively, which we interpret to
293 result from the release of stored elastic strain energy (Supplementary Figure. 7b and c).

294 Samples prepared using Ar ion milling were taken from 3mm cores drilled into hand specimens.
295 These cores were then encased in brass tubes using crystalbond adhesive and sliced into ~0.5 mm
296 thick discs with a low powered diamond bladed saw. The discs were then reduced to a thickness of
297 ~100 µm by polishing with 600 and 1200 grit papers on a water based automatic polisher and
298 finished to a high quality using 2500 and 4000 grit papers on the same machine. The polished 100
299 µm thick discs were removed from their protective brass rings and cleaned of crystalbond by
300 immersion in acetone for several hours. Finally, selected discs were placed in a Gatan precision ion
301 polishing system (PIPS) and milled with dual Ar ion beams until a perforation was formed within or
302 bordering a biotite grain. Heterogeneous milling rates, due to the multiphase nature of the samples,
303 complicated this process, with the first perforation sometimes not occurring in a biotite grain,
304 meaning that further milling was required.

305 **TEM analysis**

306 Transmission electron microscopy experiments were performed using a CEOS GmbH “CESCOR”
307 probe side aberration corrected JEOL 2100FCs instrument, operating at 200keV. The resolution
308 offered by the microscope is 1.4Å using conventional TEM illumination and sub-Ångstrom using
309 aberration corrected STEM.

310 Low resolution imaging and EDX measurements were performed using a tungsten filament JEOL
311 2000FX TEM operating at 200keV. Extra care was taken during TEM analysis due to the nature of
312 micas as beam sensitive materials and the fact that the delamination features of interest were
313 initially hypothesized to be a result of beam damage. Beam exposure was kept to a minimum in
314 areas of interest; alignments and focusing were carried out in sacrificial areas of the sample and
315 critical regions exposed only during image capture. Some areas were imaged at multiple
316 magnifications meaning they were exposed to the beam for longer. In these instances, beam
317 damage was often observed but as described in the main text, this took the form of amorphisation
318 and a loss of contrast. The expansion structures on which this study is focused were not observed to
319 grow or form during extended exposure to the beam except in some regions in close proximity to
320 the perforation edge of PIPS prepared samples.

321 Chemical analysis including beam induced K loss measurements, were performed using an EDAX
322 energy dispersive X-ray (EDX) detector attached to the JEOL 2000FX TEM. Previously unexamined
323 regions of biotite were selected for measurements to ensure no prior diffusion of K from beam
324 exposure. Spectra were taken from target regions using collection times of either 90 live seconds or
325 120 live seconds, an amp time constant of 102.4 μ s, detector area of 30mm² and a detector energy
326 resolution of 136eV at FWHM Mn k-alpha. Measurements were repeated at the same location using
327 a constant live time to investigate the loss of K between each spectrum. During this period the beam
328 was not moved from its position to ensure all spectra were collected from the same spot. Spectra
329 were mathematically filtered to remove background data. K/Si ratios were used as described by Van
330 der Pluijm et al.³⁵ so as to normalise the loss of K to that of Si.

331 **Data availability**

332 The data associated with this study are available from the corresponding author upon request.

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431 phyllosilicate-bearing fault gouges. *J. Geophys. Res. Solid Earth* **112**, 1–12 (2007).

432

433 End Notes

434

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441

442 Author contributions:

443 JA collected samples, prepared specimens, produced and analysed TEM data and wrote the
444 manuscript. EM supervised the work, collected samples, provided guidance in image interpretation,
445 data analysis and geological implications and helped write the manuscript. KD prepared FIB samples,
446 produced TEM data and provided instruction and guidance on TEM analysis and interpretation.
447 MWB provided guidance on ripplocation theory and interpretation of microstructures. All
448 contributed to improvements of the manuscript.

449

450 Competing interests

451 The authors have no competing interests to declare.

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456

457 Figure Legends

458

459 **Figure 1. Phyllosilicate structures and ripplocation morphology.** (a) – (c) Atomic structure models of
460 some common phyllosilicates with valences identified by polygons. The indices of these polygons are
461 occupied by O atoms or OH groups. (a) Biotite, like other micas, requires an interlayer cation (I) to
462 balance the overall negative charge of its tetrahedral (T) and octahedral (M) layers. In the cases of
463 biotite and muscovite this cation is predominantly K whereas in paragonite, and others, it is Na. (b)
464 Lizardite, like the other serpentine minerals antigorite and chrysotile has a 1:1 ratio of T to M layers
465 which possess a neutral charge resulting in no interlayer cations. (c) The structure of chlorite is
466 similar to biotite except that the interlayer cations are replaced by individual octahedral layers, so
467 called brucite like sheets. (d) and (e) Comparison between a dislocation (d) and a ripplocation in a
468 generic phyllosilicate lattice (e). Both enable the movement of one plane of atoms over another.
469 However, while dislocation climb is required to deform out of this plane in (d), ripplocations
470 intrinsically contain an element of c-axis parallel strain due to bending and more crucially are
471 attracted to each other rather than repelled.

472 **Figure 2. Diamond shaped expansion structure arrays.** Brightfield TEM micrographs of FIB-prepared
473 biotite specimens from mylonitic orthogneiss. (a) Bright streaks parallel to the basal plane, but
474 stacked in two principle orientations, produce elongated, diamond-shaped arrays across the entirety
475 of biotite grains. The acute angles of these diamonds measure between 30° and 60°. (b) At higher
476 magnifications, these streaks are revealed to be up to 60 Å wide, lozenge-shaped lattice expansion
477 (or delamination) structures of the biotite basal planes. (c) Detail of expansion
478 structures/delaminations stacked in en-echelon arrays. (d) Schematic of the features observed in (a),
479 (b) and (c) showing the en-echelon stacking of lozenge shaped expansion structures/delaminations
480 offsetting TMT layers and producing diamond shaped arrays.

481

482 **Figure 3. Lattice curvature and interlayer expansion.** (a) Lattice curvature is visible at the scale of
483 individual TMT layers which bend up to 30° within as little as 3 nm forming ripples (indicated by
484 arrows in bottom left). Inset shows detail of the region indicated by the attached arrow overlain with
485 a biotite atomic structure model showing how the lattice expansion occurs within the interlayer. (b)
486 Profile integrated from the region defined in (a) showing a widening of brighter contrast interlayers
487 within the regular 1 nm lattice spacing of pristine biotite. The double peak to the right represents an
488 individual chlorite layer. (c) High magnification TEM lattice fringe image with biotite atomic structure
489 model overlain. Dark regions centre on octahedral layers, intermediate (grey) regions show
490 tetrahedral layers and light regions demonstrate the location of interlayers.

491

492 **Figure 4. Formation of conjugate arrays of expansion structures.** Analogy between (a) the Anderson
493 model of conjugate faulting and (b) a schematic representation of the conjugate arrays of expansion
494 structures and delaminations we observe in biotite. In both cases structures (faults in (a) expansion
495 structures in (b)) form in two principal orientations which are up to 60° apart and display opposite
496 shear sense. The obtuse angle between the two orientations is bisected by σ_3 in (a) and the c-axis in
497 (b). In both cases this is the orientation along which expansion (or extension) occurs. In this way the
498 diamond shaped expansional arrays are crystallographically controlled as is evidenced by the fact
499 that they have different orientations in different grains, being always aligned with the c-axis of each
500 grain. Conjugate delamination arrays represent the most efficient way to relieve stored grain-scale c-
501 axis strain energy, just as conjugate normal faults result from extension on a larger scale. The
502 orientations of principal stresses in this figure are therefore representative of the time when stored
503 elastic strain energy is released (during thinning for TEM), not necessarily the time during which
504 strain was being accumulated (deformation within the viscous shear zone).

505 **Figure 5. Conceptual model showing how ripplocations lead to kink bands or conjugate expansion**
506 **structure arrays.** A pristine undeformed biotite lattice (a) experiences differential stress with some
507 component of compression parallel to the basal planes. As the lattice is confined this induces
508 numerous small bulk ripplocations which store the resulting c-axis parallel strain energy (b).
509 Continued basal plane parallel compression imparts enough energy for ripplocations on different
510 layers to migrate and merge to form a kink band between two kink band boundaries of opposite sign
511 (c). The elastic c-axis strain energy becomes permanent strain on the formation of mature through-
512 going kink bands. This may produce the kinked microstructures common in micas from viscously
513 deformed rocks. Parts of a biotite lattice that have experienced a stress field similar to (b) but in
514 which kinking has not been induced still contain stored elastic c-axis strain energy in the form of
515 ripplocations. This energy is released on thinning for TEM analysis in the form of conjugate arrays of
516 expansion structures and delaminations which facilitate a grain-scale c-axis parallel expansion (d).
517 This is represented by the structures shown in Fig. 2 of this study.