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**Experimental observation of carousel-like phason flips in the
decagonal quasicrystal $\text{Al}_{60}\text{Cr}_{20}\text{Fe}_{10}\text{Si}_{10}$**

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22 Quasicrystals show long-range order, but without translational symmetry^{1,2}.
23 Unexpectedly, we directly observed the carousel-like successive flipping of groups of
24 atoms inside the ~ 2 nm decagonal structural subunits of the decagonal quasicrystal
25 $\text{Al}_{60}\text{Cr}_{20}\text{Fe}_{10}\text{Si}_{10}$ using *in situ* high-temperature high-resolution transmission electron
26 microscopy imaging. The observed directionally successive phason flips occur mainly
27 clockwise and occasionally anticlockwise. The origin of these directional phason flips
28 is analyzed and discussed.
29

30 Generally, two or more unit tiles are utilized to describe the crystal structures of
31 quasicrystals, where the tiles have to abide by strict matching rules in the case of
32 perfect quasiperiodic tilings. However, local phasonic disorder has been widely found
33 experimentally^{3,4}. Quenched phasons are generally reflected by shifts and broadenings
34 of diffraction spots in the diffraction patterns or by jagged atomic rows in
35 high-resolution transmission electron microscopy (HRTEM) images³. Recently, we
36 found a quasicrystal-related mosaic with aperiodically distributed unit tiles inlaid in
37 the lattice of translationally periodic structural blocks, where the origin of this
38 distinctive feature was also ascribed to phasonic disorder⁵. The actions of phason flips
39 were experimentally observed indirectly by neutron scattering⁶⁻⁸, Moessbauer
40 spectroscopy⁹, and NMR^{10,11}. Importantly, Edagawa *et al.* carried out the first direct
41 observations of phason flips in an Al-Cu-Co decagonal quasicrystal (DQC) using *in*
42 *situ* high-temperature HRTEM^{12,13}, where the phason flips were corresponding to the
43 rearrangements of the unit tiles by a few atomic jumps. They observed
44 thermally-induced go-and-return transitions between A and B positions in the
45 Al-Cu-Co DQC¹². Trebin *et al.* related the observability of phason flips in HRTEM
46 experiments, which are on the timescale of seconds to minutes, to the atomic flip rate
47 which is of the order of picoseconds¹⁴. The growth of decagonal quasicrystals was
48 also found to be related with phason-flip-related error-and-repair processes instead of
49 following strict local growth rules¹⁵. In addition, successive phason flips involving
50 more than two positions were also observed in the Al-Cu-Co DQC¹³. However, these
51 successive phason flips were generally considered as random actions because the

52 occupied positions of flipping atoms in the tiling were at random and no evident rules
53 were obeyed. Besides, thermally-induced atomic fluctuations/atomic jumps were
54 observed in other systems, where the routes of atomic jumps are also random¹⁶. These
55 facts raise the question of the possibility of regular correlated dynamical flips of
56 atoms or clusters.

57 In this paper, we report peculiar thermally fluctuating phasons in decagonal
58 $\text{Al}_{60}\text{Cr}_{20}\text{Fe}_{10}\text{Si}_{10}$, where a carousel-like successive flipping of atoms is observed
59 directly using *in situ* high-temperature HRTEM. We found that the successive
60 clockwise or occasionally anti-clockwise phason flips of atoms are located on a 0.77
61 nm decagon inside the ~ 2 nm decagons. This is the first time, as far as we know, that
62 a carousel of successive phason flips could be observed. The origin for this novel kind
63 of phason flips is analyzed and discussed in the following.

64

65 **Results and discussion**

66 Figure 1a shows a selected-area electron diffraction pattern (EDP) of decagonal
67 $\text{Al}_{60}\text{Cr}_{20}\text{Fe}_{10}\text{Si}_{10}$ at 800°C, where the diffraction spots are arranged tenfold
68 symmetrically (for example, those marked by circles), characteristic for DQCs. Figure
69 1b presents a typical HRTEM image viewed along the ten-fold axis of the DQC at
70 800°C; the features of the DQC are reproduced by the corresponding fast Fourier
71 transform in Fig. 1c, where the marked spots correspond to those marked in Fig. 1a.
72 An enlarged HRTEM image from the thin area of Fig. 1b, overlaid with a proper tiling,
73 is shown in Fig 1d. Its unit tiles mostly correspond to stars (S), boats (B), hexagons

74 (H), bowties (BT) and, occasionally, to decagons (D) with almost perfect tenfold
75 symmetry. Note that the combination of B, H, BT tiles may further form decagons
76 with a diameter of ~ 2 nm, as can be seen in Fig. 1e. This kind of D clusters can be
77 classified in two types: one consists of two H and one B tile (highlighted in purple),
78 another one of three H and one BT tile (highlighted in yellow). For clarity, these two
79 typical D clusters are denoted as D_{2H+1B} and D_{3H+1BT} , respectively. For comparison,
80 the structural features of decagonal $Al_{60}Cr_{20}Fe_{10}Si_{10}$ at room temperature (RT) are also
81 shown in the atomic-resolution high-angle annular dark field scanning transmission
82 electron microscopy (HAADF-STEM) image in Figs. 1f, g. Enlarged images of the
83 two typical D clusters, D_{2H+1B} and D_{3H+1BT} , are depicted in Figs. 1h and i. There, the
84 vertices of the unit tiles are occupied by smaller decagonal atomic clusters with a
85 diameter of ~ 0.47 nm. The different imaging conditions of HRTEM and
86 HAADF-STEM do not allow a direct comparison of atomic arrangements at high and
87 at low temperature. However, the unit tiles are the same at both temperatures,
88 implying that the vertices of structural tiles in Fig. 1d and e represent the positions of
89 columnar atomic clusters. We emphasize that the D clusters with broken tenfold
90 symmetry in decagonal $Al_{60}Cr_{20}Fe_{10}Si_{10}$ consist of the unit tiles H, B, or BT, which
91 was also shown previously^{17,18}. Therefore, the change of the structural units inside a \sim
92 2 nm D cluster will be a rearrangement among the H, B, or BT tiles.

93 Thermally-induced atomic phason flips with directionally successive transitions,
94 discovered using *in situ* high temperature HRTEM, are shown in Figs. 2-4 (could be
95 also found in the Supporting Video 1). The phason flips are confined on the decagon

196 with a diameter of 0.77 nm within the larger ~ 2 -nm D clusters. Interestingly, these
197 special atomic phason flips occur mainly along the clockwise direction, which was
198 not expected. We take three highlighted D clusters, designated as D1, D2 and D3 in
199 Fig. 2a, as examples to demonstrate the directionally successive transitions of
100 thermally-induced atomic phason flips.

101 Firstly, we focus on the change of D1 in Figs. 2a-f, where the time is measured
102 from the moment of Fig. 2a and with the elapsed times of 0, 1, 2, 3, 4, and 5 s,
103 respectively. We note that the directionally successive transitions occur inside the D1,
104 as schematically summarized in the first row of Fig. 2g. We use the same color to
105 denote the H tiles with the same orientation. The vertices of the subtiles inside the \sim
106 2-nm D cluster could be some out of 10 spots on the 0.77 nm circle, as marked as 1,
107 2, ... 10 in the first \sim D cluster in the first row of Fig. 2g. Which unit tiles form where
108 depends on which one of the 10 marked positions in the first D cluster in Fig. 2g are
109 occupied. However, the motif made by two occupied positions with the interval of
110 one empty position on the 0.77 nm D cluster is geometrically forbidden because it
111 would bring a geometric conflict for the generation of the observed structural tiles of
112 DQC (see Supplementary Fig. 2 in Supporting Materials).

113 As seen clearly from the first row of Fig. 2g, the changes are limited to the inner
114 spots of the ~ 2 -nm D1 cluster, and not to the ten spots at the periphery of 2-nm D1
115 cluster. The detailed processes of directionally dynamical phasons are analyzed in the
116 lower part of Fig. 2g, where the changeable positions are indicated with the
117 corresponding numbers. Starting from the first circle in the bottom row of Fig. 2g

118 (corresponding to the inner circle with a diameter of 0.77 nm in the D1 in Fig. 2c), the
119 occupied positions 2, 5, 8 and 9 at 2 s are flipped to the positions of 2, 3, 6 and 9 at 3 s,
120 and then to 3, 4, 7 and 10 at 5s. It is evident that the position 5 at 2 s continuously
121 flips to the next position, 6, at 3s and then to 7 at 5s, forming a continuous clockwise
122 motion (as summarized in Fig. 2h). After these flips took place, the atoms on the 0.77
123 nm decagon inside the D1 cluster are mainly located on the positions 3, 4, 7, 10, with
124 occasionally flipping to the neighboring positions (not shown here).

125 Such a carousel of successive atomic jumps is also found in the D2 cluster, but it
126 occurred at a different time compared to that in the D1 cluster, as seen in Fig. 3. Note
127 that the measured time of Fig. 3 is based on the frames in Fig. 2a. There are two
128 groups of directionally successive flipping of atoms along clockwise direction, and
129 each group involves four changeable positions. Interestingly, both groups proceed
130 synchronously: they start at the positions 2 and 9 at 71 s, and then continuously hop to
131 the following positions at 72 s, 73 s, and 74 s, respectively, as analyzed in Fig. 3e as
132 well as summarized in Fig. 3f and in the table at the bottom-right corner of Fig. 3,
133 where the two groups of continuous flips are indicated by arrows with different colors.
134 However, no successive flipping of atoms is observed in the nearby D1 cluster for the
135 moment.

136 In addition to the clockwise successive flipping of atoms, anticlockwise
137 successive phason flipping is also occasionally observed, as seen in the D3 tile in Fig.
138 4. There are three groups of directionally successive atomic jumps in the D3 tile, as
139 summarized in Fig. 4f, where two of them are clockwise and one is anticlockwise.

140 We now analyze the anticlockwise dynamical flipping from the position 10 to 9,
141 and finally to 8 in the D3 tile. When the atoms at position 3 at 28 s flip to the position
142 2 at 29 s, the atoms at position 10 at 28 s have to flip to the position 9 at 29 s to
143 maintain a physically reasonable geometric configuration, namely to retain two, rather
144 than one, empty interval positions between two occupied positions (as discussed
145 above). Meanwhile, the atoms at position 10 cannot flip to the position 1 because it
146 would generate a structural block not found in Al-Cr-Fe-Si DQCs (see Supplementary
147 Fig. 2 in Supporting Materials). In the next step, the atoms at position 2 at 29 s flip
148 back to the position of 3 at 30 s, performing a generally observed go-and-return
149 transition^{12,13}. The atoms at position 9 at 29 s are maintained at 30 s. In the next
150 moment, the atoms at position 3 at 30 s flip to the position 4 at 31 s, forming a
151 clockwise successive phason flipping from position 2 to 3, and finally to position 4.
152 Meanwhile, another group of clockwise successive phason flipping from the position
153 4 to 7 proceeds simultaneously. When the atoms at position 6 at 30 s flip to the
154 position 7 at 31 s, the atoms at position 9 at 30 s flip to the position 8 at 31 s to
155 maintain a physically reasonable geometric configuration, namely to eliminate the
156 forbidden configuration with one interval position between two occupied positions on
157 the 0.77 nm D cluster. Consequently, anticlockwise successive phason flipping from
158 position 10 to 9 and finally to 8 takes place, accompanying two groups of successive
159 flipping of atoms along clockwise direction, as summarized in Fig. 4f and the table
160 right side of Fig. 4f.

161

162 **Conclusions**

163 We observed experimentally a carousel of successive flipping of thermally fluctuating
164 phasons in decagonal $\text{Al}_{60}\text{Cr}_{20}\text{Fe}_{10}\text{Si}_{10}$ by using *in situ* high temperature HRTEM
165 imaging. The atomic positions of directionally successive phason flipping were
166 located on the small decagons with a diameter of 0.77 nm, inside larger ~ 2 -nm
167 decagons. Successive phason flipping took place mainly clockwise and occasionally,
168 only, anticlockwise. We found that this special kind of phason flipping is closely
169 related with the geometric constraints of reasonable structural blocks in DQCs.
170 However, the underlying mechanisms need to be explored theoretically as well in the
171 future. We believe that this experimental finding might excite a wide interest in
172 revealing its underlying mechanisms and finding its potential applications.

173

174 **Methods**

175 **Preparation of the quasicrystals**

176 Approximately 1 kg of the master alloy with a nominal composition $\text{Al}_{60}\text{Cr}_{20}\text{Fe}_{10}\text{Si}_{10}$
177 was first prepared by melting the high-purity elements in an induction furnace under
178 an argon atmosphere. The molten alloy was then poured into a graphite crucible in the
179 furnace to form an ingot. Some fragments of the obtained ingot were heated at 1100°C
180 for 7 days in an evacuated quartz tube, and then cooled in the furnace by switching off
181 the power.

182 **Characterizations**

183 The atomic-resolution structural investigations were performed with a
184 JEM-ARM200F microscope equipped with a cold field emission gun, Cs-probe
185 corrector, and Cs-image corrector. *In situ* HRTEM observations were carried out in a
186 Thermo Fisher Titan environmental transmission electron microscope equipped with
187 an image aberration corrector and operated at 300 kV. The crushed alloys were
188 diluted in ethanol and dispersed with ultrasonic treatment. The solution was then
189 dripped on a Protochips Fusion *in situ* heating chip, which was then mounted on a
190 double-tilt Protochips Fusion TEM heating holder. The heating history of the sample
191 is shown in Supplementary Fig. 1 in the supporting material. The shape of the
192 observed sample was produced by electron irradiation at elevated temperatures.
193 HRTEM images at 800°C were taken at a magnification of 3.8×10^5 and recorded by
194 a Gatan US1000 CCD camera, using a Gatan video recording software, with a
195 frame-rate of 1 frame per second.

196 **Data availability**

197 The data that support the findings of this study are available from the corresponding
198 author upon reasonable request.

199

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208 Slovak Academy of Sciences, Slovakia.

209

210 **Author contributions**

211 Z.H. conceived the research; Z.H. and J-L.M. performed the experiments; Z.H., W.S.
212 and J-L.M. wrote the manuscript., Z.H., J-L.M., H.M., H.L., T.Z., Y.W., X.M., and
213 W.S. analysed the data, discussed the results, and drew the conclusions.

214

215 **Competing interests**

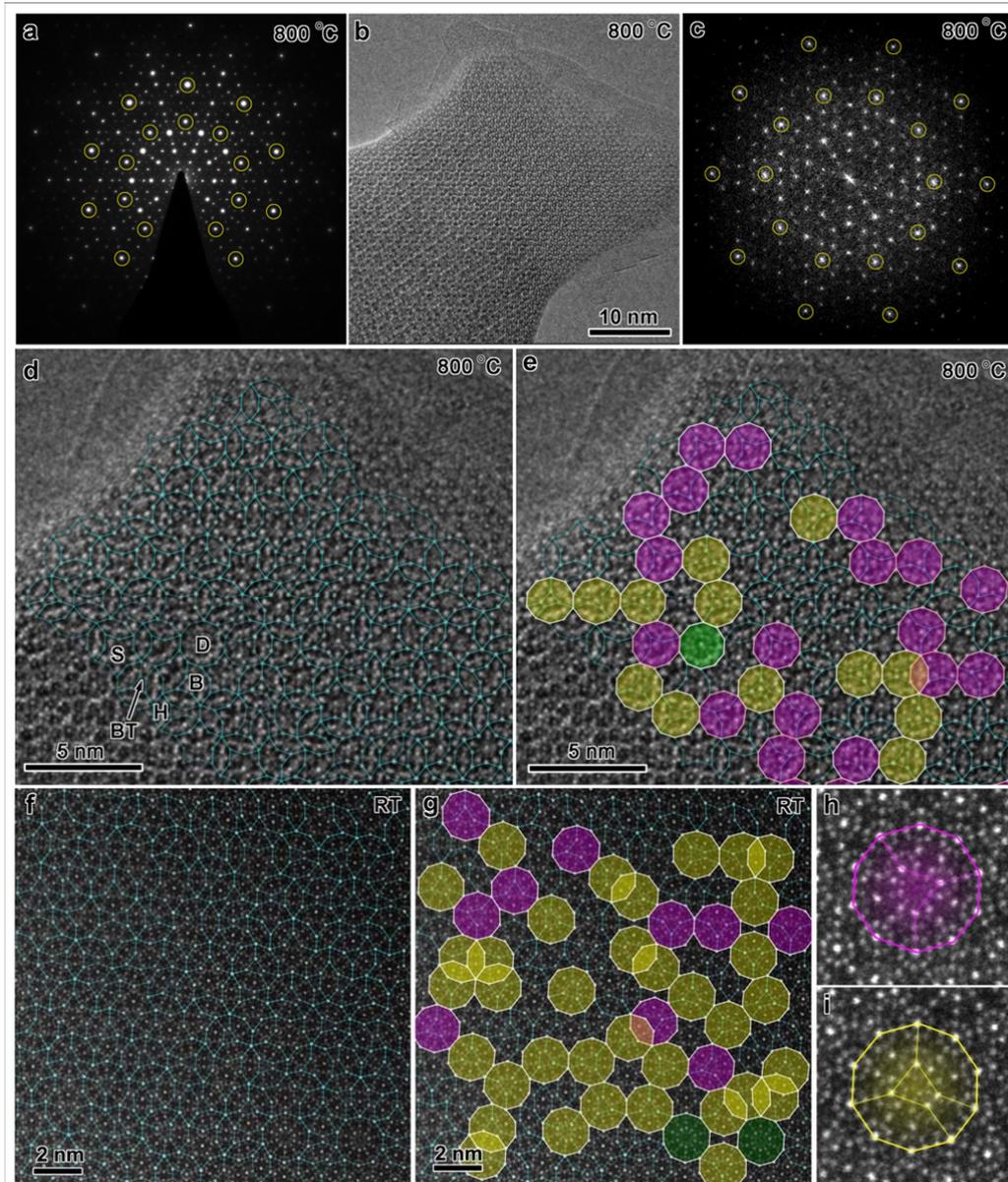
216 The authors declare no competing interests.

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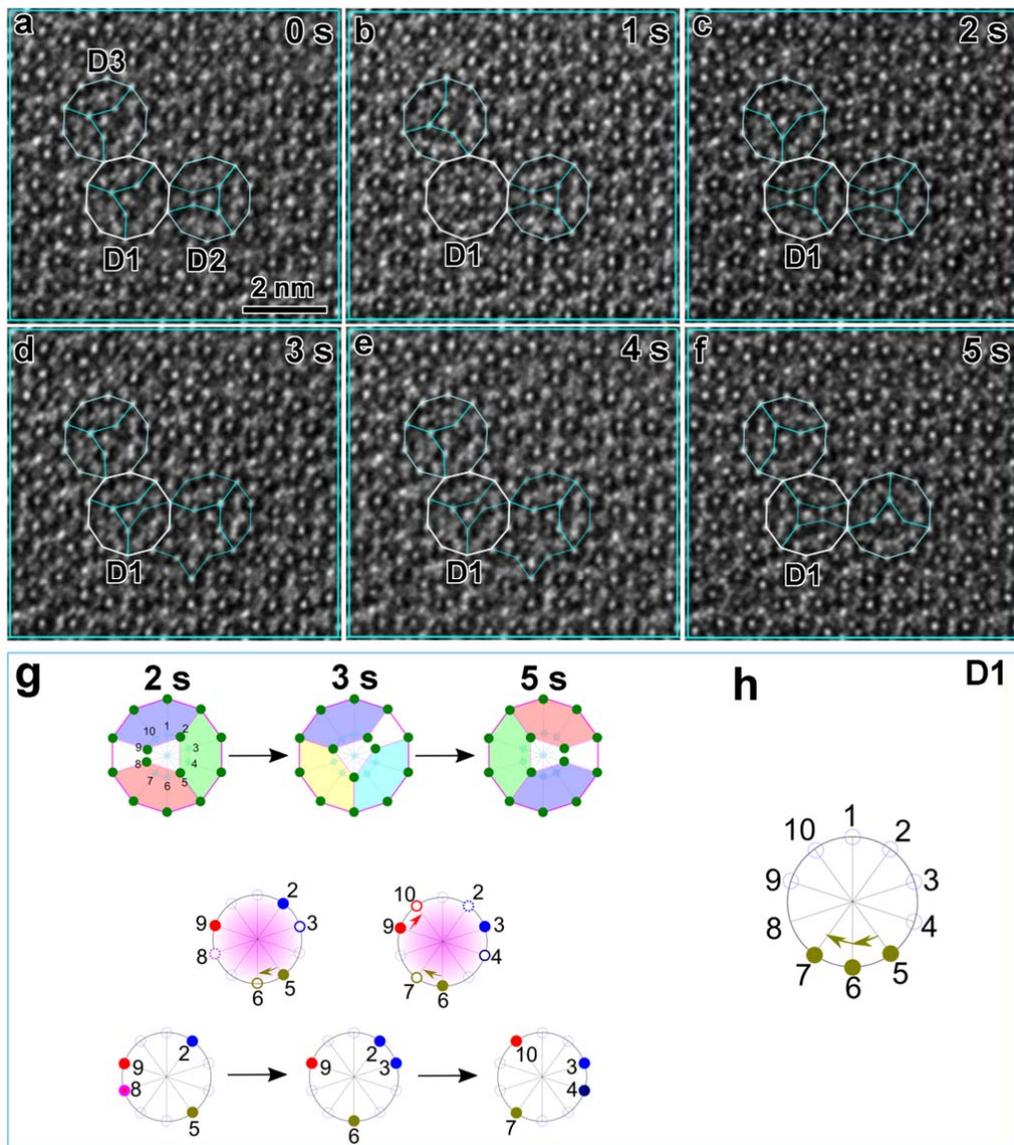


263

264 **Fig. 1| Basic structural information of decagonal $\text{Al}_{60}\text{Cr}_{20}\text{Fe}_{10}\text{Si}_{10}$ viewed along**
 265 **the tenfold zone axis. a,** Selected-area electron diffraction pattern of the DQC taken
 266 **at 800°C.** The diffraction pattern exhibits an evident tenfold symmetry, as shown by
 267 the yellow circles. **b,** HRTEM image of the DQC observed at 800°C. **c,** the FFT of **b,**
 268 which also exhibits the features of the DQC. **d** and **e,** Enlarged HRTEM images of the
 269 DQC at 800°C with the structural blocks depicted by polygons **e,** and with the ~2-nm
 270 D clusters emphasized (the same color means the same type of D cluster). **f** and **g,**

271 Atomic-resolution HAADF-STEM image of DQC with the structural blocks depicted
272 in polygons **f**, and with the ~ 2 -nm D clusters emphasized in **g**. The same color means
273 the same type of D cluster. **h** and **i**, Enlarged image of two typical D clusters: D_{2H+B} in
274 **h**, D_{3H+BT} in **i**.

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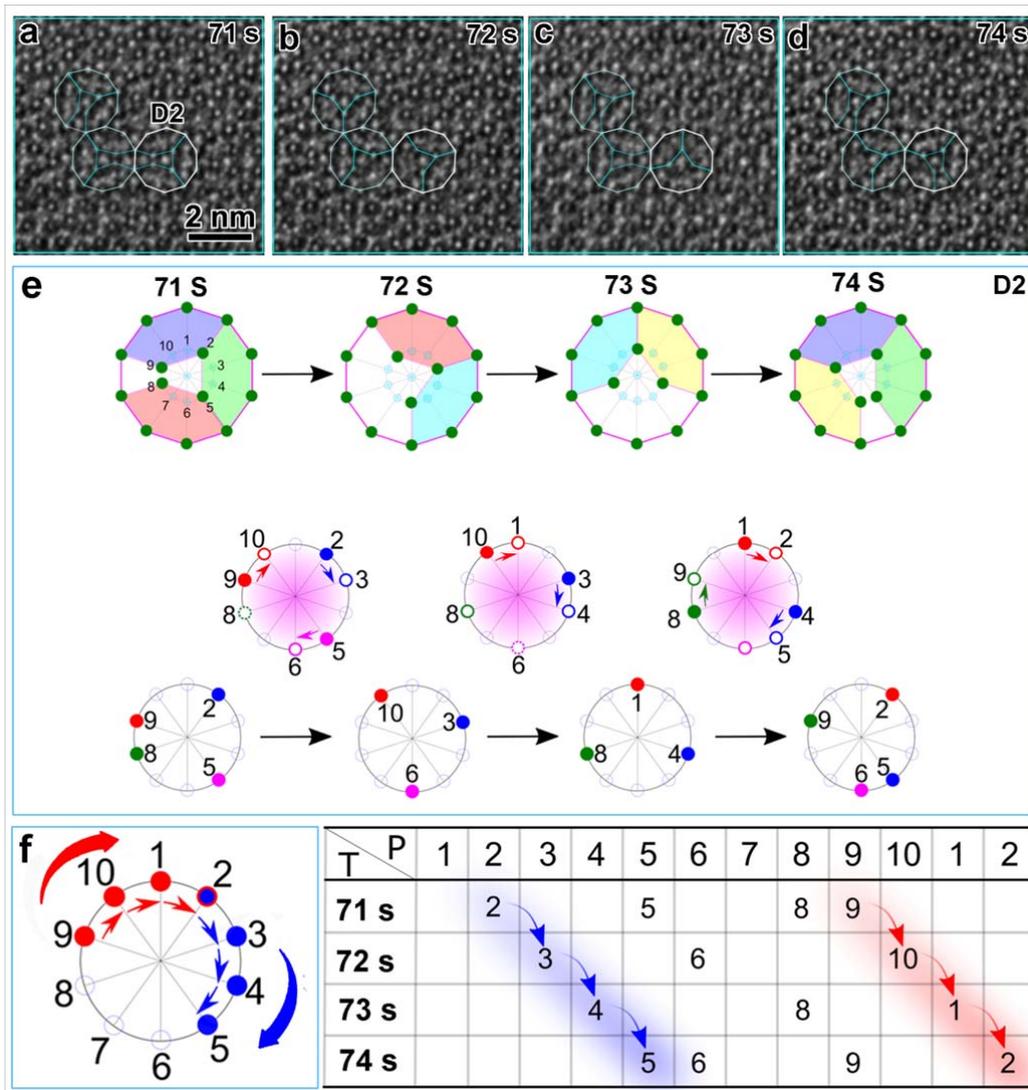


277

278 **Fig. 2| Successive clockwise propagation of atomic phason flips inside of the D1**
 279 **cluster at 800°C. a-f, HRTEM images of the DQC taken at different times. Three**
 280 **~2-nm D clusters are marked, but we focus on the changes of the D1 cluster, as**
 281 **highlighted in white outline. g, Geometric schematic of the structural changes of D1**
 282 **as time going on. In the first row, the ten numbers of 1, 2, ...10 correspond to positions**
 283 **of possible atomic columns inside the first ~2-nm D cluster, located on a smaller**
 284 **circle with a diameter of 0.77 nm. The positions occupied by atomic columns are**
 285 **indicated by green spots. In the middle and bottom row, the changes of atomic**

286 positions on the 0.77 nm D cluster are summed up. The occupied positions are
287 indicated by solid spots and marked by numbers. The hopping of atomic columns is
288 marked by arrows in the middle row. **h**, Summary of continuous clockwise flips of
289 atoms inside the D1 cluster.

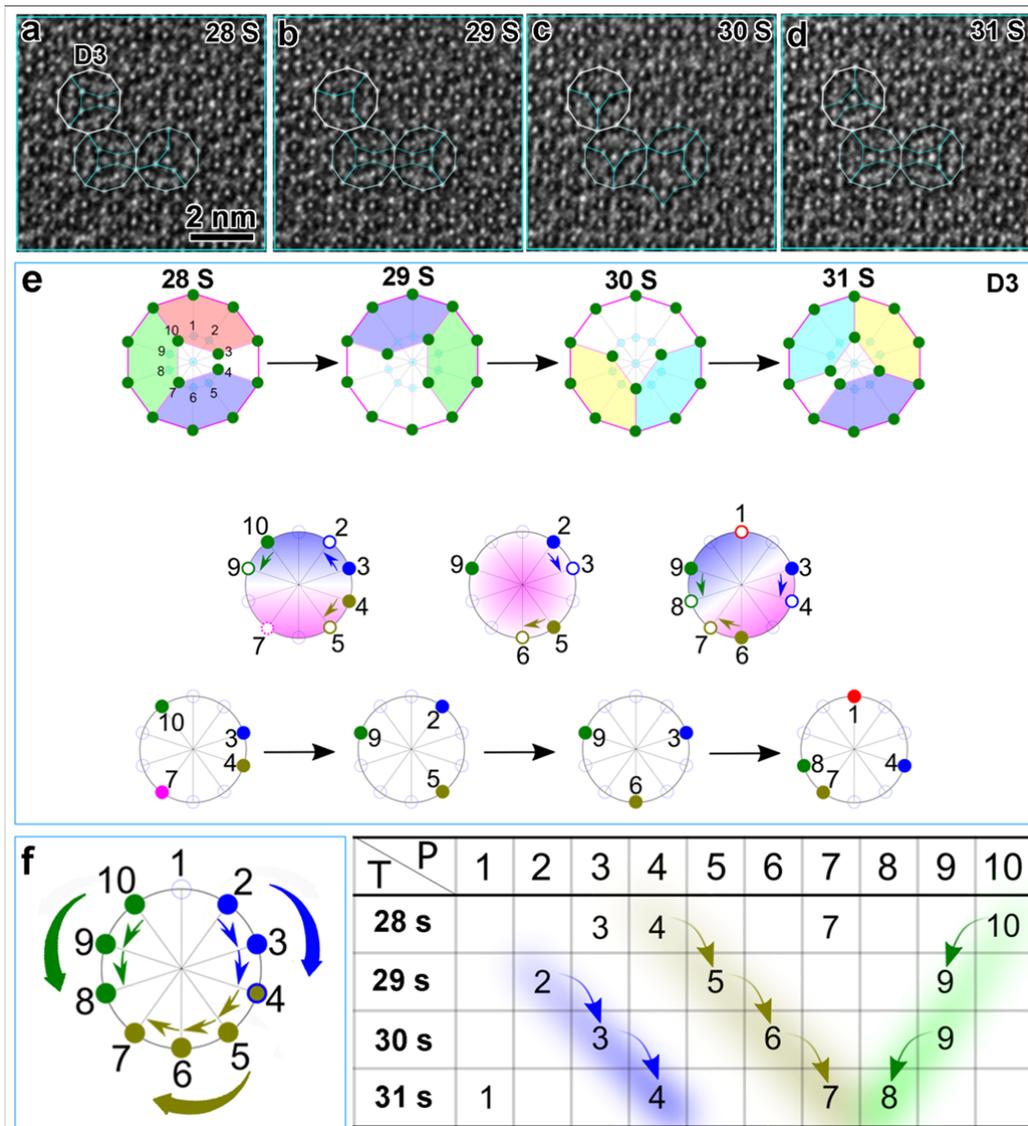
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292

293 **Fig. 3| Successive clockwise propagation of atomic phason flips inside of the D2**
 294 **cluster at 800°C. a-d,** HRTEM images of the DQC taken at different times. Three
 295 ~ 2 -nm D clusters are marked, but we focus on the change of the D2 cluster, as
 296 highlighted in white outline. **e,** Geometric schematic of the structural change of D2 as
 297 time is going on. As in Fig. 2, the ten numbers of 1, 2, ...10 mark possible atomic
 298 column positions in a smaller circle with a diameter of 0.77 nm. The positions
 299 occupied by atoms are indicated by green spots. In the middle and bottom row, the
 300 changes of atomic positions on the 0.77 nm circle are summarized. The occupied
 301 positions are indicated by solid spots and marked by numbers. The atom movements

302 are marked by arrows in the middle row. **f**, Summary of two groups of continuous
303 clockwise flips of atoms in D2 cluster. The arrows indicate the direction of dynamical
304 processes and the colors of arrows represent different groups. The "P" in the table at
305 the bottom-right corner means the "position" occupied by atoms on the 0.77 nm circle
306 inside the ~2 nm D cluster; and "T" means time.
307



309

310 **Fig. 4| Successive clockwise propagation of atomic phason flips inside of the D3**
 311 **cluster at 800°C. a-d**, HRTEM images of the DQC taken at different times. Three
 312 \sim 2-nm D clusters are marked, but we focus on the change of the D3 cluster, as
 313 highlighted in white outline. **e**, Geometric schematic of the structural change of D3 as
 314 time is going on. (same presentation as in Figs 2 and 3). **f**, This time, contrary to the
 315 cases in Figs. 2 and 3, hopping is observed in both clockwise and anticlockwise
 316 directions.