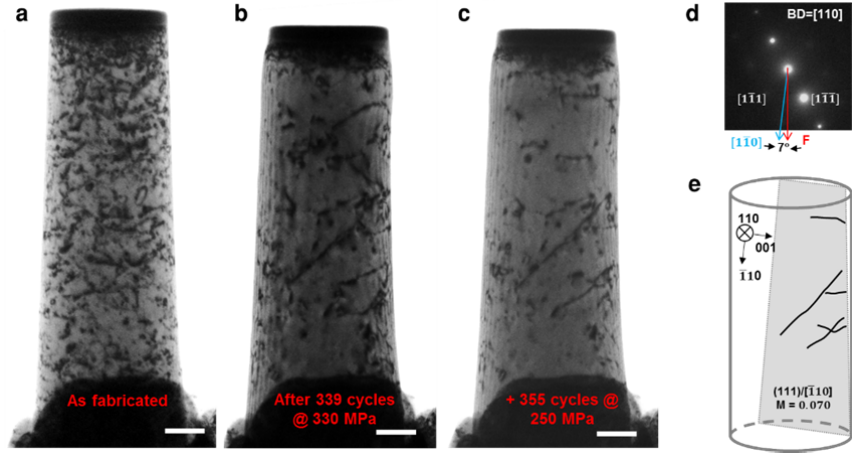
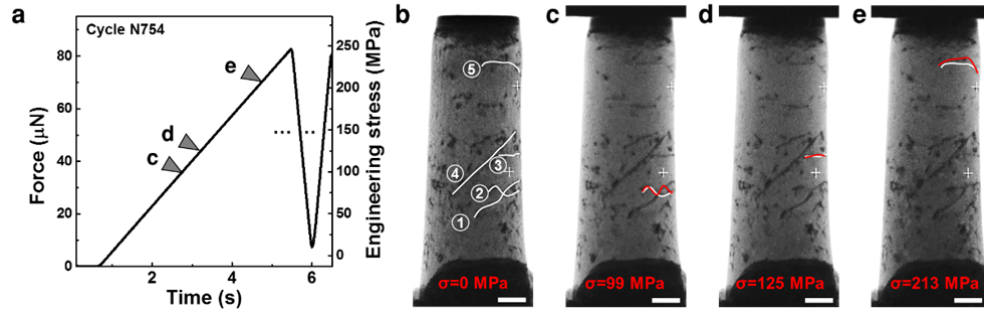
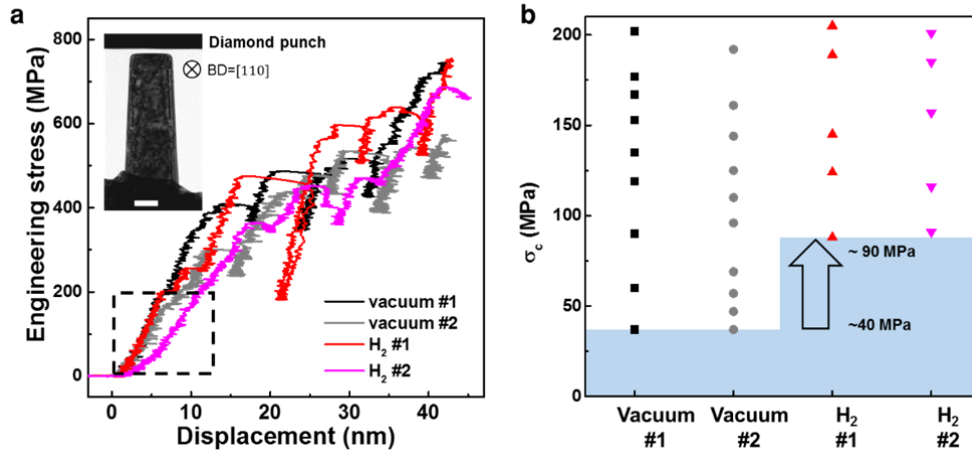




Supplementary Figure 1. Process to achieve individual-dislocation configuration through small-strain-amplitude cyclic compression. (a) As-fabricated pillar contains high density of dislocations. (b) After 339 cycles of cyclic compression with peak stress of 330 MPa, the dislocation density dramatically decreased. (c) After another 355 cycles of cyclic compression with peak stress of 250 MPa, only a few long dislocations remained inside the pillar. (d) The diffraction pattern of the single crystal aluminum pillar, for which the loading axis is inclined to the $[1\bar{1}0]$ direction by 7° . (e) Schematic of the mobile dislocations in (c) and one of the possible slip system, for which the Schmidt's factor M is calculated to be 0.070. All scale bars represent 200 nm.



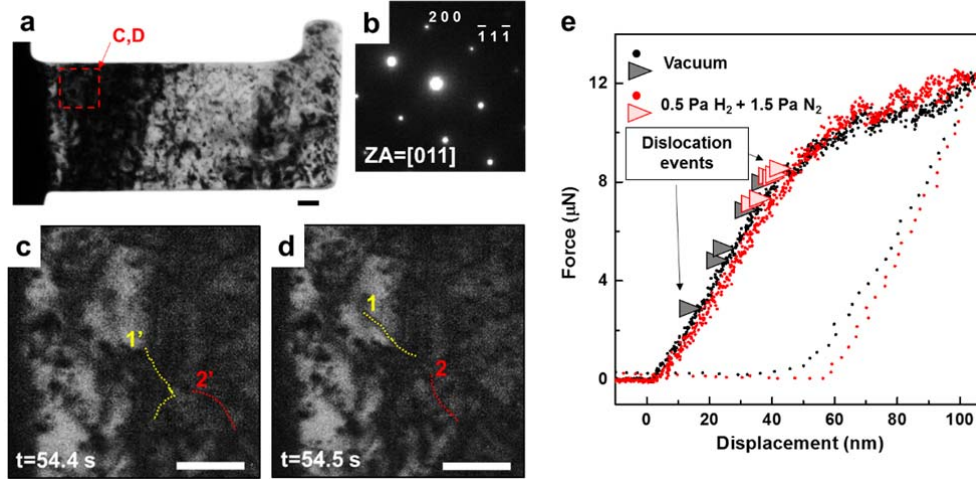
Supplementary Figure 2. The threshold loads for each dislocation determined in the first loading cycle in vacuum. (a) Load vs. time curve of the slow leading cycle. The triangular markers correspond to the threshold loads of each dislocation to be activated. (b) The original configuration of the dislocations in the pillar before loading. (c), (d), and (e) show the abrupt jump of dislocations #2, #3 and #5 at critical loading stresses of 99, 125, and 213 MPa respectively. The red lines and white lines are the dislocation profiles before and after the jump. For more details see supplementary Movie 1. All scale bars represent 200 nm.



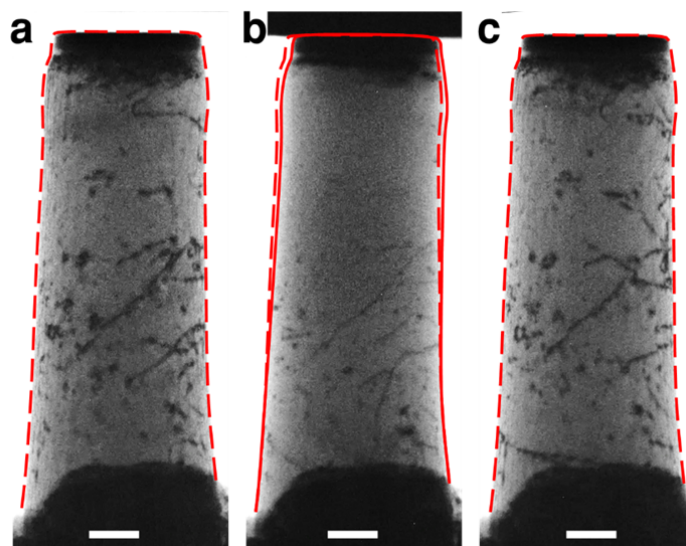
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22 **Supplementary Figure 3. Monotonic compression of aluminum pillars in vacuum and**
 23 **hydrogen gas. (a)** Engineering stress-displacement curves showing similar yield stress and flow
 24 stress for tests in vacuum and in hydrogen. The inset illustrates the experimental setup. **(b)** The
 25 critical stress (σ_c) for dislocation depinning events in various samples, showing that a higher stress
 26 is required to set off dislocation motion in hydrogenated samples. Scale bar in inset in (a) is 200
 27 nm.

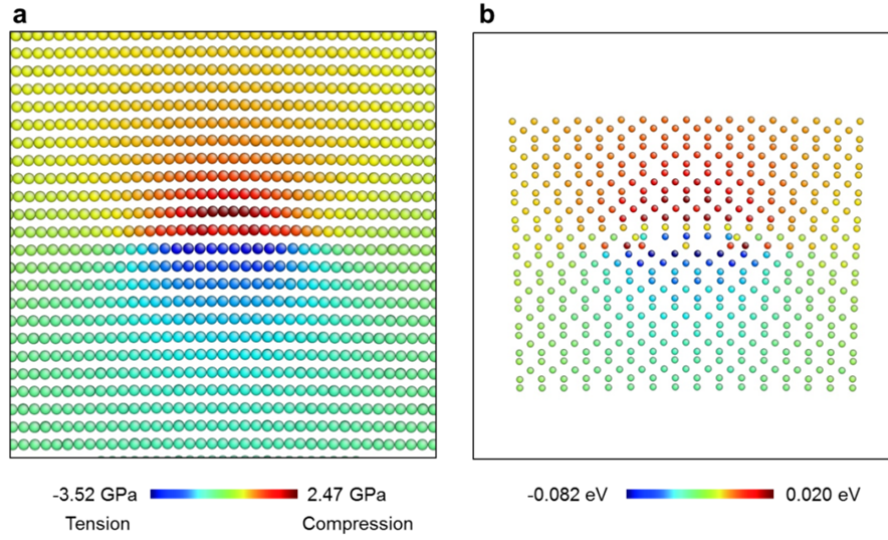
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Supplementary Figure 4. Hydrogen effect on dislocation motion during bending tests. (a) Bright field TEM image of a typical bending cantilever containing dislocations and other defects. (b) The corresponding diffraction pattern showing the single crystal nature of the cantilever. (c) and (d) are the magnified area in (a) showing two typical dislocation profile at the given moments before and after sudden movements. (e) Force vs. displacement curves of two typical samples with similar dimensions but tested in vacuum and hydrogen environment, respectively. The first 5 dislocation events of each test are labeled on the curve with triangle marks. All scale bars represent 100 nm.



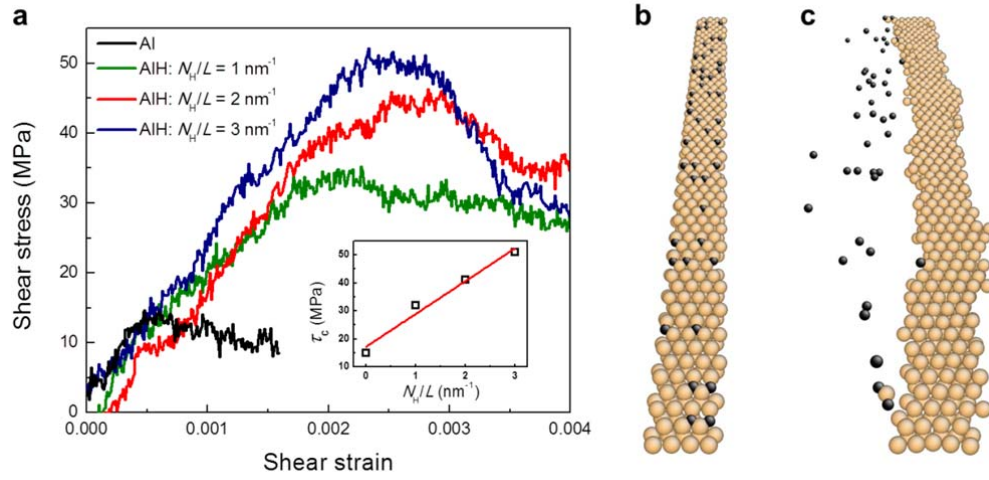
Supplementary Figure 5. Destruction of dislocation configuration due to buckling during the test in 2 Pa H₂. (a) The dislocation configuration inside the pillar remained unchanged after 85 cycles. (b) Due to the tip drift, the pillar was buckled. The dislocation configuration underwent a sudden change under the coupling of loading stress and buckling stress. Note pillar contour change before drift (red dashed line) and after drift (red solid line), as well as the change of black contrast inside the pillar due to buckling. (c) When the tip is detached from the pillar, the pillar contour recovered but the dislocation configuration can't be resumed. All scale bars represent 200 nm.



52

53 **Supplementary Figure 6.** The distribution maps of (a) hydrostatic stress of edge dislocation in
 54 aluminum and (b) total energy difference of hydrogen ($\Delta E_{H,i}$) in interstitial sites around the
 55 dislocation core. Generally, interstitial hydrogen in the region with minus hydrostatic stress
 56 (tension) has a smaller value of $\Delta E_{H,i}$ and that in the region with positive hydrostatic stress
 57 (compression) has a larger value of $\Delta E_{H,i}$. The tensile region around dislocation core corresponds
 58 to the most energy-favored sites for hydrogen atoms.

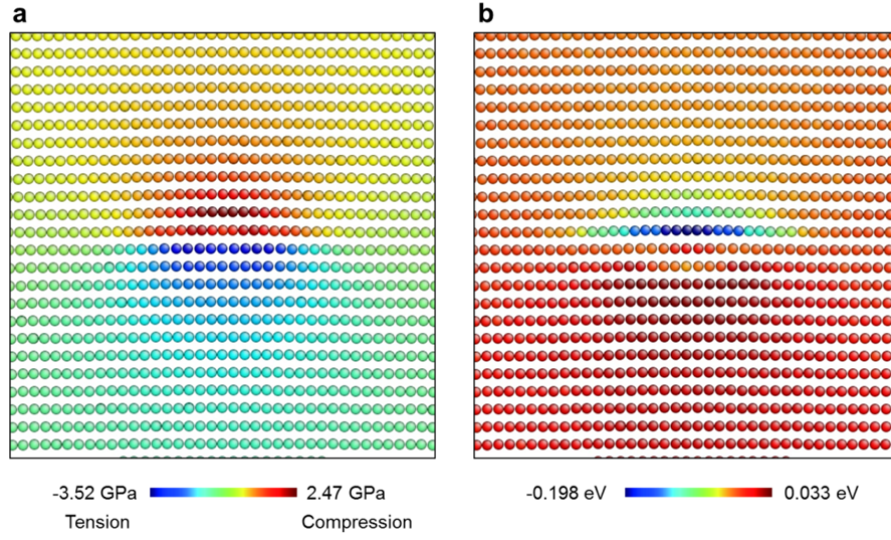
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60

61 **Supplementary Figure 7. Atomistic simulations based on the model with hydrogen atoms in**
 62 **a Boltzmann distribution. (a)** Stress-strain curves at different hydrogen concentration levels. The
 63 critical shear stress (τ_c) for dislocation motion shows a linear increase with the increase of
 64 hydrogen concentration (N_H/L), as shown in the inset. **(b)** Initial configuration of dislocation
 65 decorated with hydrogen atoms. **(c)** The configuration at the depinning state. Atoms with golden
 66 and black colors refer to aluminum and hydrogen.

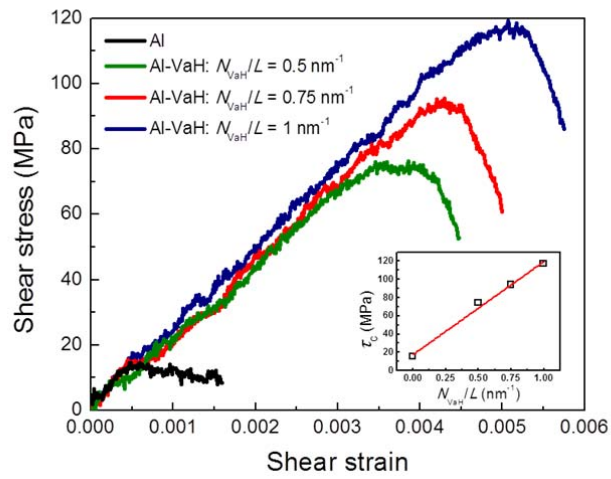
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68

69 **Supplementary Figure 8.** The distribution maps of (a) hydrostatic stress of edge dislocation in
 70 aluminum and (b) total energy difference of VaH complex ($\Delta E_{\text{VaH},i}^{\text{Edge}}$) in site i ($E_{\text{VaH},i}^{\text{Edge}}$) with
 71 reference to that in bulk site ($E_{\text{VaH},i}^{\text{Bulk}}$).

72



73

74 **Supplementary Figure 9. MD simulations of dislocation pinning effect by hydrogen-vacancy**
 75 **complex.** The different colored lines show the stress-strain curves at different concentrations of
 76 hydrogen-vacancy complex. The critical shear stress (τ_c) for dislocation motion shows a linear
 77 increase with the increase of hydrogen-vacancy complex concentration (N_{VaH}/L), as shown in the
 78 inset.

79

80 **Supplementary Table 1.** Energetics of isolated H atom in Al. ΔE_s refers to the solution energy of
81 H in Al. T and O refer to the tetrahedral (T) interstitial and octahedral (O) interstitial sites. E_m
82 refers to the migration barrier of one H atom from a T-site to the O-site. The units are in eV.

	ADP	ab initio
ΔE_s (T)	0.69	0.69 (ref. 1)
ΔE_s (O)	0.82	0.82 (ref. 1)
E_m	0.19	0.18 (ref. 1)

83

84

85

86 **Supplementary Table 2.** Energetics of Va in Al. E_v^f refers to the vacancy formation energy and E_m
87 refers to the vacancy migration barrier. The units are in eV.

	ADP	Experiments
E_v^f	0.68	0.69 (ref. 2)
E_m	0.64	0.65 (ref. 3)

88

89

90

91 **Supplementary Table 3.** Energetics of VaH. E_f (T-Va) refers to the formation energy of vacancy
92 with H in T-site. E_f (O-Va) refers to the formation energy of vacancy with H in O-site. E_b (T-Va)
93 refers to the binding energy of vacancy with H in T-site. The units are in eV.

	ADP	ab initio
E_f (T-Va)	1.32	1.0 (ref. 4)
E_f (O-Va)	1.36	1.14 (ref. 4)
E_b (T-Va)	0.048	0.39 (ref. 5)

94

95

Supplementary Note 1: Mechanical annealing by cyclic compression

The loading function consists of a slow leading cycle and tens of fast trailing cycles in each group of test. In the loading half-cycle, each dislocation marches forward, like a bending bow, with both ends pinned on surface; upon unloading, each swings back to the original location (see supplementary Movie 1).

The initial compression tests were conducted with peak stress of ~330 MPa. By the end of 339th cycle (N339), only a few isolated mobile dislocations were left, with their both ends pinned by the surface of the sample (Supplementary Fig. 1b). In order to stabilize these dislocations further, we purposely lowered the peak stress down to ~250 MPa. A steady dislocation configuration was reached at N694 (Supplementary Fig. 1c), i.e. for each following cycle, the bow out extent of each dislocation remained the same at each peak stress.

In Supplementary Fig. 2, three dislocations show abrupt position change during the loading, whereas the two other dislocations only show smooth position change. Critical loading stress for dislocation #1, #3, and #5 are 99 MPa, 125 MPa, and 213 MPa respectively.

Supplementary Note 2: Monotonic compression

Four typical engineering stress-strain curves from monotonic compression tests are shown in Supplementary Fig. 3. Before the stress reaches 200 MPa, the curve looks linear without obvious yielding or strain burst. But when correlated with the movie frames, discrete movements of preexisting dislocations can be observed. These preexisting dislocations in the as-fabricated pillars mainly result from FIB damage in the milling process, and are pinned by obstacles such as surface junctions, point defects and other dislocations. As the applied load increases, dislocations start breaking away from pinning points, exhibiting abrupt movements. After depinning,

122 they either vanish at surfaces or get stopped at a stronger pinning point. We have
123 tracked all the observed depinning events and their corresponding critical stresses, and
124 found that the lowest depinning stress increases from ~40 MPa to ~90 MPa when
125 changing the testing environment from vacuum to the 2 Pa hydrogen gas, as
126 demonstrated in Supplementary Fig. 3b.

127 From the engineering stress-displacement curves, monotonic compressions in
128 hydrogen environment show similar stiffness and strain hardening behavior when
129 compared to compressions in vacuum. In these tests, the profile of stress-strain curve
130 is mainly governed by sample-size dependent dislocation nucleation and
131 multiplication. In Supplementary Fig. 3a, H₂ #2 specimen show lower stiffness than
132 the other three pillars due to the slight misalignment and initial point contact at the
133 beginning of the compression test.

134

135 **Supplementary Note 3: Hydrogen effect on dislocation motion in bending tests**

136 8 smooth cantilevers with width ranging from 222 nm to 292 nm and thickness from
137 600 nm to 642 nm were bent inside the ETEM. All the cantilevers (e.g.
138 Supplementary Fig. 4a) contained preexisted dislocations resulting mainly from FIB
139 milling. The dislocation density was estimated to be between 10^{13} m^{-2} and 10^{14} m^{-2} .
140 Monotonic bending loads were applied to the cantilevers at loading rate of 5 nm/s
141 under displacement control mode. Total 8 samples were tested, 4 in vacuum and 4 in
142 hydrogen environment. Similar to that observed in monotonic compression tests
143 (Supplementary Fig. 3), the threshold loads for dislocation activation in the hydrogen
144 atmosphere are always higher than those tested in vacuum. One typical comparison is
145 shown in Supplementary Figure 4e. It again demonstrates that hydrogen can improve
146 the threshold loads significantly for those preexisted dislocations.

147

148 **Supplementary Note 4: Reconfiguration of dislocations due to buckling/bending**
149 **induced by probe drift**

150 Since a typical cyclic loading test lasts 40 to 100 seconds, the diamond punch may
151 undergo obvious drift in all three space coordinate directions. Under the load control
152 system, drift in the indentation direction can be automatically compensated by the
153 feedback control system so as to ensure a correct loading force applied to the pillar.
154 However, drifts perpendicular to the indentation axis can buckle the samples
155 obviously due to the friction confinement at the contact interface between the
156 diamond tip and the samples. This will apply additional stress to the dislocations. One
157 typical example is shown in Supplementary Fig. 5. The sample was bended/buckled
158 obviously (Supplementary Fig. 5b) during the cyclic loading, as suggested by the
159 profile deviation (red dashed lines) and the significant contrast change. New
160 dislocations coming out from the bottom disrupted the dislocation configuration
161 inside the pillar. Interestingly, the sample can go back to its original position after the
162 diamond probe was retracted despite that the dislocation configurations had been
163 dramatically changed.

164

165 **Supplementary Note 5: Evaluation of Al-H angular dependent potential**

166 To provide more details about the empirical angular-dependent potential (ADP)⁶ we
167 used in MD simulations, here we show various properties predicted by this potential
168 in comparison to the experiments and ab initio calculations, especially some
169 energetics related to the point defects (H, Va and VaH), as shown in the following
170 three Tables 1, 2 and 3. Among them, most has a relatively good match with the
171 referenced data, but unfortunately, the binding energy of VaH is very small. More
172 information for this potential is shown in refs 6, 7.

173

174 **Supplementary Note 6: Molecular dynamics simulation of the influence of**
175 **hydrogen atmosphere on dislocation motion**

176 We first created the $a/2\langle 110 \rangle$ -type edge dislocations in Al. The simulation box was
177 oriented along x -[10 $\bar{1}$], y -[111], z -[1 $\bar{2}$ 1] , with dimensions of 23nm $\times$ 28nm $\times$ 16nm.
178 Periodic boundary conditions were applied in the x and z directions. The hydrogen
179 atoms were then introduced in the interstitial tetrahedral sites around the dislocation
180 according to the Boltzmann occupation probability

$$181 \quad p_{H,i} = e^{\Delta E_{H,i}/k_B T} / \sum_i e^{\Delta E_{H,i}/k_B T} \quad (S1)$$

182 where $\Delta E_{H,i} (= E_{H,i} - E_{H,i}^{\text{Bulk}})$ is the total energy difference of hydrogen in site i ($E_{H,i}$)
183 with reference to that in bulk tetrahedral interstitial site ($E_{H,i}^{\text{Bulk}}$), k_B is Boltzmann
184 constant and T is absolute temperature. Supplementary Fig. 6a shows the distribution
185 of hydrostatic stress of the edge dislocation and Supplementary Fig. 6b shows the
186 distribution of $\Delta E_{H,i}$ in tetrahedral site around the dislocation core. We can see that
187 the two maps match relatively well. The tensile region corresponds to a much lower
188 energy state, where most of the hydrogen atoms distribute. The most energy-favored
189 sites for hydrogen are right in the dislocation core region. The hydrogen concentration
190 around dislocation line is characterized with N_H/L , where N_H is the number of
191 hydrogen atoms and L is the length of the dislocation line⁸. After adding hydrogen
192 atoms, the system was further relaxed at 300 K for 100 ps using a Nosé-Hoover
193 thermostat^{9, 10}. Finally shear strain was applied in the xz -plane to drive dislocation
194 glide along the x direction. The strain rate was on the order of 10^8 s^{-1} . The whole
195 calculations were carried out using the LAMMPS code¹¹ and the atomic
196 configurations were displayed via AtomEye¹².

197 Supplementary Fig. 7a shows the shear stress-strain curves for Al-H system in
198 comparison to H-free system at 300 K. An obvious locking effect can be observed: the

value of critical shear stress for edge dislocation motion increases in hydrogen environment. The dependence of locking strength on hydrogen concentration is studied by conducting a series of simulations with different N_H/L . The inset in Supplementary Fig. 7a shows the variation of critical shear stress (τ_c) with different amount of N_H/L . We find that the value of τ_c increases almost linearly with hydrogen concentration as

$$\tau_c = \tau_0 + \alpha N_H / L \quad (\text{S2})$$

where τ_0 (= 15 MPa) is the critical shear stress for dislocation motion without hydrogen and the coefficient α is 11.7 MPa·nm in the present Al-H system. It is interesting to observe that even hydrogen with a density of $N_H/L = 1 \text{ nm}^{-1}$ could double the τ_c . Supplementary Fig. 7b shows the initial configuration of dislocation decorated with hydrogen atoms at the level of $N_H/L = 3 \text{ nm}^{-1}$. When the shear stress reaches the critical value, the dislocation suddenly escapes from the hydrogen atmosphere, as illustrated in Supplementary Fig. 7c.

Our MD simulations reveal in a straightforward manner that hydrogen atoms could enhance the critical stress for dislocation motion. The interaction between dislocation and diffusing solute hydrogen atoms can lead to locking effect, similar to that in strain aging^{13, 14}.

217

218 **Supplementary Note 7: Molecular dynamics simulation of the influence of**
219 **hydrogen-vacancy complexes on dislocation motion**

Our simulations above show that the hydrogen atoms themselves can take the pinning effect on dislocation motion. However, as we mentioned in the manuscript, the aging experiments show that the relocking needs a long time beyond hydrogen diffusion time. It is reasonable to speculate that there are some other obstacles to lock dislocations. By calculating the migration energy and estimating the diffusion length, we found that the most probable candidate is hydrogen-vacancy complex. In the

following, another set of simulations was performed to reveal the role of hydrogen-vacancy complex on dislocation motion. Similar to that in Al-H system, we introduced the VaH complexes according to the Boltzmann occupation probability

$$p_{\text{VaH},i} = e^{\Delta E_{\text{VaH},i}/k_B T} / \sum_i e^{\Delta E_{\text{VaH},i}/k_B T} \quad (\text{S3})$$

where $\Delta E_{\text{VaH},i}$ is the total energy difference of VaH complex in site i ($E_{\text{VaH},i}$) with reference to that in bulk site ($E_{\text{VaH},i}^{\text{Bulk}}$). Supplementary Fig. 8a shows the distribution of hydrostatic stress of the edge dislocation and Supplementary Fig. 8b shows the distribution of $\Delta E_{\text{VaH},i}$ around the dislocation core. Supplementary Fig. 9 shows the shear stress-strain curves at different concentrations of hydrogen-vacancy complexes. It can be seen that the more hydrogen-vacancy complexes are implanted, the stronger pinning effect will be. The critical shear stress (τ_c) for driving dislocation motion also shows a linear response with the line density of hydrogen-vacancy complex (N_{VaH}/L), but with a much steeper slope (101.9 MPa·nm) than that pinned by hydrogen. Our current simulations show that the hydrogen-vacancy complex take an even stronger pinning role than the hydrogen atmosphere.

241

242

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