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Supporting Information

An Organoferroelasticity Driven by Molecular Conformational Change

Sajjad Husain Mir^a, Yuichi Takasaki^{a,b}, Satoshi Takamizawa^a*

^a Department of Materials System Science, Graduate School of Nanobioscience, Yokohama City

University, 22-2 Seto, Kanazawa-ku, Yokohama, Kanagawa 236-0027, Japan

^b Kanagawa Institute of Industrial Science and Technology, Shimoimaizumi, 705-1 Ebina, Kanagawa, 243-0435, Japan

*E-mail: staka@yokohama-cu.ac.jp

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Other supplementary material

Video files for bending action of 1 observed under a polarizing microscope:

(a) Mechanical twinning at 298 K:	Takamizawa_movS1.mov
(b) Stress-Strain test at 298 K:	Takamizawa_movS2.mov

(a) Experimental information

i) Stress-strain test

Stress tests were carried out on a universal testing machine (Tensilon RTG-1210, A&D Co. Ltd.).

ii) Single-crystal X-ray diffraction experiment

Single-crystal X-ray analysis of 1 was performed at 298 K on a Bruker SMART APEX CCD area (graphite-monochromated Mo-K α radiation ($\lambda = 0.71073$ Å) with a nitrogen flow temperature controller. In the measurement, a bent crystal of 1 was used as shown in Figure S2. Empirical absorption corrections were applied using the SADABS program. The structure was solved by direct methods (SHELXS-97) and refined by full-matrix least squares calculations on F^2 (SHELXL-97) using the SHELX-TL program package. Non-hydrogen atoms were refined anisotropically; hydrogen atoms were refined in a riding model. The crystal face indexing was carried out using SMART in a SHELXTL Ver.6.12 program package with a twin resolution program. Crystallographic data of the structure is summarized in Tab. S1.

(b) Crystallographic data



Figure S1. Molecular structures of **1** in (a) mother domain and (b) twinned domain as ORTEP representations drawn at 50% probability level for the ellipsoid obtained from single crystal X-ray diffraction measurement at 298 K.

Domain	Mother	Twinned
T/K	298	298
Empirical formula	C ₆ H ₁₀ O ₄	C ₆ H ₁₀ O ₄
Crystal size /mm ³	0.26×0.14×0.13	0.26×0.14×0.13
М	146.14	146.14
Crystal system	Monoclinic	Monoclinic
Space group	P2 ₁ /c	<i>P</i> 2 ₁ /c
a /Å	7.3663(12)	7.3693(14)
b /Å	5.1555(8)	5.1566(10)
c /Å	10.1436(13)	10.1453(16)
α /°	90.00	90.00
β/°	112.329(9)	112.354(11)
γ /°	90.00	90.00
V /Å ³	356.34(9)	356.55(11)
Ζ	2	2
D _{calcd} /Mg m ⁻³	1.362	1.361
μ(Mo Kα) / mm ⁻¹	0.115	0.115
Reflections collected	2510	2515
Independent reflections (R_{int})	877(0.0404)	881(0.0379)
Goodness of fit	1.031	1.035
$R_1(I > 2\sigma \text{ (all data)})$	0.0527(0.0997)	0.0547(0.1100)
$_{\rm w}R_2(I > 2\sigma \text{ (all data)})$	0.1262(0.1445)	0.1258(0.1488)
Largest diff. peak (hole) /eÅ ³	0.152(-0.132)	0.167(-0.170)

 Table S1. Crystallographic data of 1 in bent shape.

		α_0 domain	1		
(a)	X	У	Z	U(eq) ^(b)	
O(1)	9742(2)	7459(3)	8712(2)	69(1)	
O(2)	7951(2)	4000(3)	8681(2)	67(1)	
C(1)	5528(3)	4658(4)	5776(2)	49(1)	
C(2)	7068(3)	6624(4)	6583(2)	51(1)	
_C(3)	8284(3)	5897(4)	8086(2)	47(1)	

Table S2. Atomic coordinates (× 10⁴) and equivalent isotropic displacement parameters (Å²× 10³) for α_0 domain and α_1 domain.

^a Depending on the program limitations, the numbering of each atom is different from that in the main text because the asymmetric unit is a half of the molecule. It is the same for α_1 domain.

 b U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

		α_1 domain	1		
	Х	У	Z	U(eq)	
O(1)	2947(2)	-993(3)	8677(2)	65(1)	
O(2)	4741(2)	2465(3)	8712(2)	69(1)	
C(1)	527(3)	-348(5)	5776(2)	48(1)	
C(2)	2068(3)	1619(4)	6580(2)	49(1)	
C(3)	3290(3)	896(4)	8087(2)	46(1)	

Table S3. Bond lengths [Å] and angles [°] for α_0 domain and α_1 domain.

α_0 domain				
O(1)-C(3)	1.299(2)			
O(2)-C(3)	1.221(2)			
C(1)-C(1)#1	1.510(4)			
C(1)-C(2)	1.510(3)			
C(2)-C(3)	1.493(3)			
C(1)#1-C(1)-C(2)	113.1(2)			
C(3)-C(2)-C(1)	114.88(16)			
O(2)-C(3)-O(1)	122.60(18)			
O(2)-C(3)-C(2)	123.37(17)			
O(1)-C(3)-C(2)	114.03(17)			

Symmetry transformations used to generate equivalent atoms: #1 -x+1,-y+1,-z+1

α_1 domain				
O(1)-C(3)	1.219(2)			
O(2)-C(3)	1.298(2)			
C(1)-C(1)#1	1.510(4)			
C(1)-C(2)	1.510(3)			
<u>C(2)-C(3)</u>	1.497(3)			
C(1)#1-C(1)-C(2)	112.7(2)			
C(3)-C(2)-C(1)	114.89(17)			
O(1)-C(3)-O(2)	123.0(2)			
O(1)-C(3)-C(2)	122.97(19)			
O(2)-C(3)-C(2)	114.05(19)			

Symmetry transformations used to generate equivalent atoms: #1 -x,-y,-z+1

Table S4. Anisotropic displacement parameters ($Å^2 \times 10^3$) for the α_0 domain and α_1 domain. The anisotropic displacement factor exponent takes the form: $-2p^2[h^2 a^{*2} U^{11} + ... + 2 h k a^* b^* U^{12}]$

			α_0 domain			
	U ¹¹	U ²²	U ³³	U ²³	U ¹³	U ¹²
O(1)	64(1)	74(1)	49(1)	11(1)	-1(1)	-21(1)
O(2)	63(1)	76(1)	46(1)	16(1)	5(1)	-18(1)
C(1)	44(1)	55(1)	41(1)	7(1)	8(1)	1(1)
C(2)	49(1)	53(1)	43(1)	8(1)	8(1)	0(1)
C(3)	41(1)	54(1)	40(1)	1(1)	10(1)	0(1)
			α_1 domain			
	U ¹¹	U ²²	U ³³	U ²³	U ¹³	U ¹²
O(1)	63(1)	76(1)	42(1)	16(1)	3(1)	-18(1)
O(2)	68(1)	73(1)	45(1)	11(1)	-2(1)	-23(1)
C(1)	45(1)	55(2)	38(1)	8(1)	8(1)	1(1)
C(2)	50(1)	54(2)	37(1)	7(1)	9(1)	1(1)

39(1)

C(3)

43(1)

52(1)

0(1)

10(1)

0(1)

		α_0 doma:	in	
(a)	X	У	Z	U(eq)
H(1)	10390	6914	9513	118(11)
H(1A)	6148	2976	5844	73(7)
H(1B)	4576	4518	6221	77(7)
H(2A)	7934	6894	6075	69(7)
H(2B)	6422	8262	6589	70(7)

Table S5. Hydrogen coordinates (× 10⁴) and isotropic displacement parameters (Å² × 10³) for the α_0 domain and α_1 domain.

^a Depending on the program limitations, the numbering of each atom is different from that in the main text because the asymmetric unit is a half of the molecule. It is the same for α_1 domain.

		α_1 domai	n	
	Х	у	Z	U(eq)
H(2)	5387	1926	9515	113(12)
H(1A)	-425	-483	6220	69(7)
H(1B)	1144	-2031	5843	68(7)
H(2A)	2932	1882	6070	53(6)
H(2B)	1424	3259	6583	71(8)

(c) Crystal phase indexing

The crystal face indexing showed deformation twinning of **1**. Shear stress formed daughter domain α_1 from α_0 in rotational twinning. The twinning interfaces are $(001)_{\alpha0}//(001)_{\alpha1}$ (or $(00-1)_{\alpha0}//(00-1)_{\alpha1}$).



Figure S2. a) Optical image of the crystal **I**, (b) crystal face indices of α_1 domain (left) and α_0 domain (right).

(d) Detail information for observation of stress-strain test

Temperature / °C	Loading	Crystal dimension		Displacement
	surface	width / µm	Thickness / µm	Velocity / µm min ⁻¹
25	(-100)	103	89.3	50

Table S6: Conditions of cyclic shear test on crystal 1, shown in Fig. 3b



Figure S3. Calculation method of a dissipated energy density. The displayed stress (σ) – strain (ε) curve is typical shape in a twinning deformation. W_{in} is the input work to proceed the deformation, $V_{\alpha 1}$ is the volume of mother domain (α_0) before changing into daughter domain (α_1) by loading, F_{obs} is the observed force in the stress-strain test, x is the displacement of the jig, S_{sample} is the cross-sectional area of the crystal sample, and l is the length of the sample from the fixing point to the contacting point of the jig.



Figure S4. Raw data of stress-strain curves measured under a condition shown in Table S6. (Green: 1st cycle with a large spike accompanying a nucleation of α_1 domain, black: 2nd cycle without a nucleation of α_1 domain.) The induced α_1 domain existing at the end of the 1st cycle was manually contracted, then the 2nd cycle started which is shown in Figure 3b of the main text.

(e) Enlarged color figures described in the main text



Figure S5. Reversible twinning deformation of **1**. a) Snapshots of the twinning on mechanical stress by tweezers; (i-iii) forward direction, (iv-vi) reverse direction under a polarizing light microscope (see Movie S1 in ESI), and (b) crystal face indices of the twinned crystal **1**. Front surface of α_0 (blue) corresponds to the back surface of the α_1 (blue), which is represented by rotating the α_0 domain about the rotational axis along $[100]_{\alpha 0}$ (green arrow illustrated above the interface).



Figure S6. Measurement of stress-strain curve. a) Snapshots of the twinning deformation of **1** upon application of shear stress: (i-iii) forward direction and (iv-vi) reverse direction; (see Movie S2 in ESI), and (b) ferroelastic hysteresis loop under shear stress at 298 K, and the value of "0" in the vertical axis is set as a center of the point of starting and reversing of displacement.



Figure S7. Partial packing diagrams of overlapping mother (α_0) and daughter domain (α_1) of **1** viewed (a) along $[100]_{\alpha 0}$ and (b) along $[010]_{\alpha 0}$. Molecules form 1D molecular chains by hydrogen bonding (indicated as dotted gray lines). The 1D chain illustrated in a lower contrast color exists by a shift of a half lattice from those of the higher contrast color. Left-side inset Figure 2b indicate the *trans* and *anticlinal* conformations in an alkyl chain. Each "R" is -CH₂-COOH. Right-side inset Figure 2 indicates an estimated rotation of –COOH groups on C6 and C1 atoms at the interface to keep the hydrogen bond linkage.