Supplementary Information

Photomechanical bending of salicylideneaniline crystals

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1. **Fig. S1.** Platelike microcrystals of enol-**1**.
2. **Fig. S2.** X-ray diffraction profile of enol-**1** microcrystals.
3. **Fig. S3.** Absorption spectra of enol-**1** microcrystals.
4. **Fig. S4.** Change of the unit cell parameters of enol-**1** crystal upon UV irradiation.
5. **Video S1.** Reversible bending of the platelike enol-**1** microcrystal (73 × 4.5 × 1.1 μm) upon alternating UV irradiation (2 s on, 5 s off) with simultaneous and continuous irradiation with visible light (>390 nm). The reversible bending was repeatable over 200 cycles. Five cycles are recorded in this video.
6. **Video S2.** Upon UV irradiation, a platelike crystal (120 × 7.1 × 1.7 μm) was able to flip a silica gel particle approximately 50-fold heavier than the microcrystal itself.
7. **Video S3.** Upon UV irradiation over the glass plate, most of the crystals bent away from the light source and the stuck crystals were stripped from the glass surface.

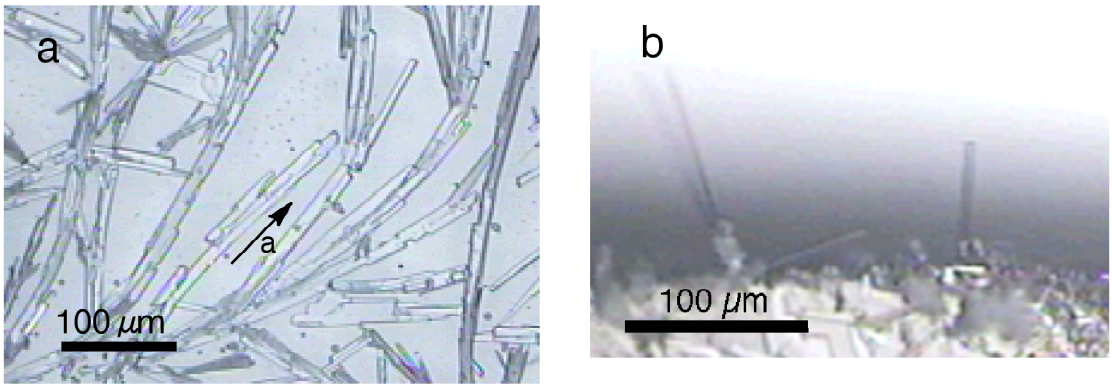

Fig. S1. Platelike microcrystals of enol-**1** grown on the surface (a) and the edge (b) of silanated glass plate by sublimation.

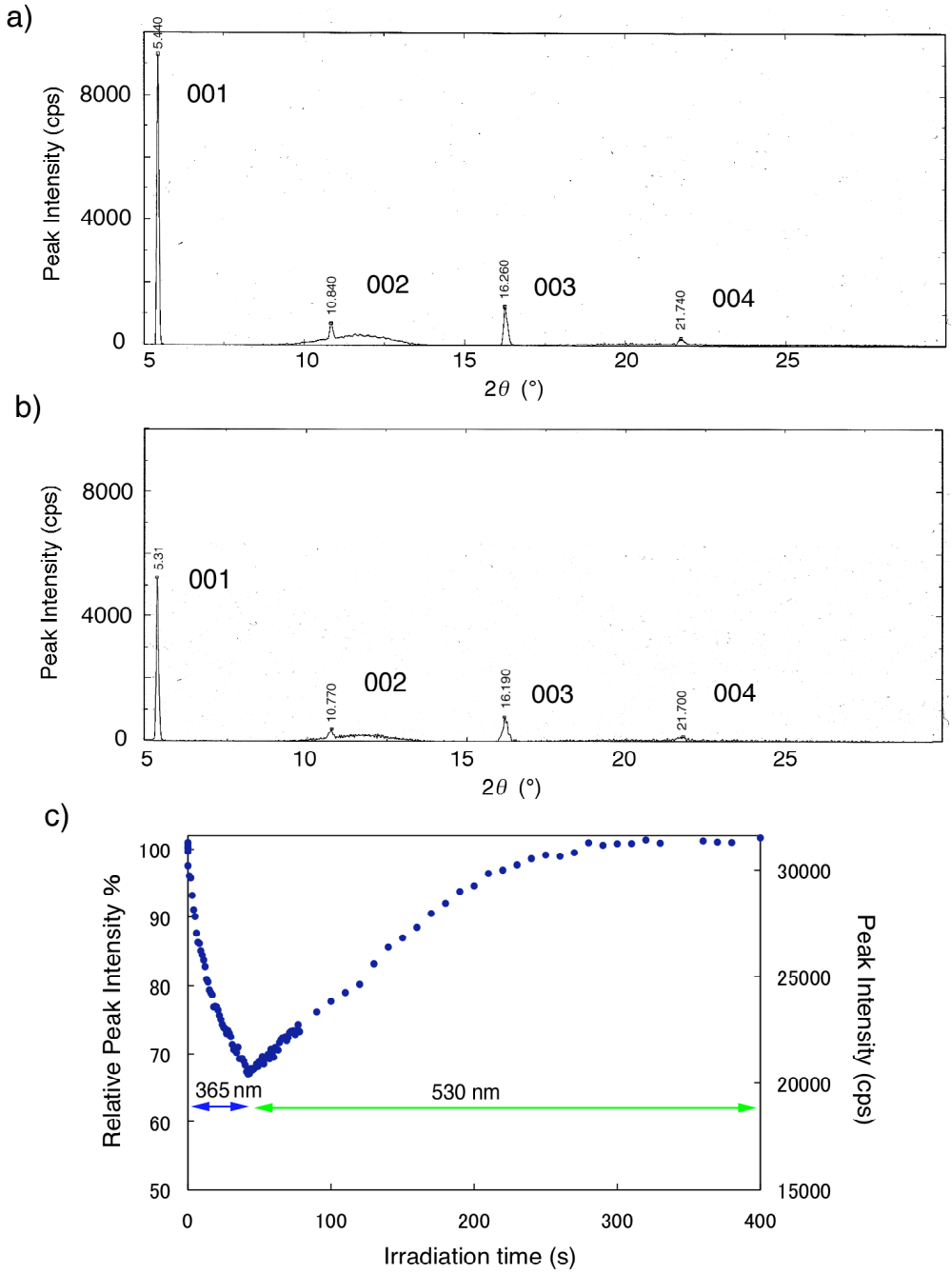

Fig. S2. X-ray diffraction profile of the enol-**1** microcrystals (a) before irradiation and (b) on maintaining UV irradiation; the intensity of peaks decreased and the positions slightly shifted to lower angle. (c) The 001 peak intensity decreased to 67% upon UV irradiation at 365 nm for 40 s and then recovered upon irradiation with visible light at 530 nm.

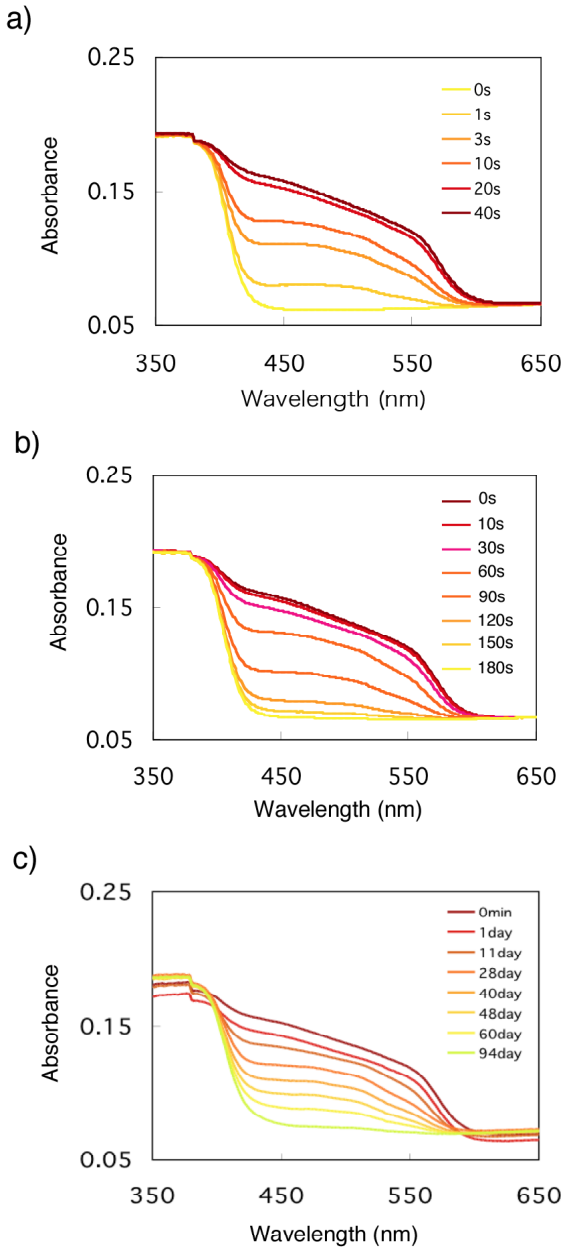

Fig. S3. Absorption spectra of the enol-**1** microcrystals upon (a) UV irradiation at 365 nm, and then (b) visible light irradiation at 530 nm, or (c) in dark on stopping UV irradiation.The lifetimes estimated from the changes of absorbance at 480 nm are (a) 2.6 s, (b) 63 s, and (c) 42 days.

UV irradiation time(s)	0	900	1800	3600	
a (Å)	6.09139(17)	6.1150(4)	6.1146(4)	6.1172(4)	**(+0.42%)**
b (Å)	10.2994(2)	10.3353(8)	10.3347(7)	10.3417(7)	**(+0.41%)**
c (Å)	16.2874(4)	16.2230(11)	16.2172(9)	16.2127(10)	**(–0.46%)**
α (deg)	102.1852(14)	102.055(4)	102.050(4)	102.017(4)	
β (deg)	92.2252(16)	92.705(4)	92.728(3)	92.797(3)	
γ (deg)	102.9942(16)	103.264(5)	103.264(4)	103.296(4)	
V (Å^3)	969.23(4)	971.06(12)	970.57(10)	971.23(11)	**(+0.21%)**

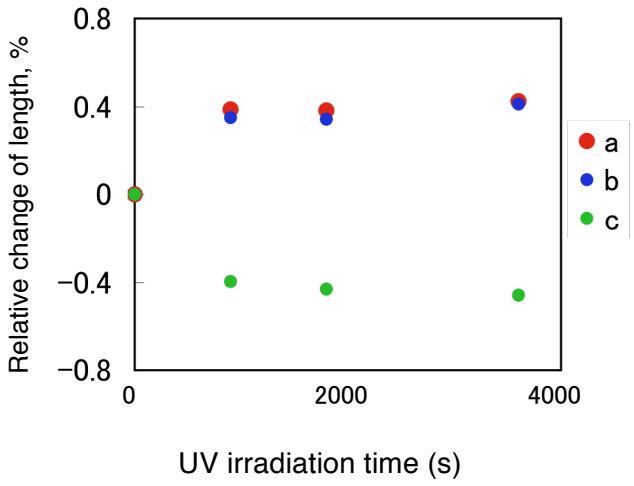

Fig. S4. Change of the unit cell parameters of enol-**1** crystal upon UV irradiation.

Video S1. Reversible bending of the platelike enol-**1** microcrystal (73 × 4.5 × 1.1 μm) upon alternating UV irradiation (2 s on, 5 s off) with simultaneous and continuous irradiation with visible light (>390 nm). The reversible bending was repeatable over 200 cycles. Five cycles are recorded in this video.

Video S2. Upon UV irradiation, a platelike crystal (120 × 7.1 × 1.7 μm) was able to flip a silica gel particle approximately 50-fold heavier than the microcrystal itself.

Video S3. Upon UV irradiation over the glass plate, most of the crystals bent away from the light source and the stuck crystals were stripped from the glass surface.