

Supporting Information

Electrical characterization of photochromic diarylethene films consisting of extraordinarily large crystallites

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1. Crystal film preparation of various DAEs

Figure S1 shows molecular structures of DAEs, which were used in the crystallization test. Table S1 summarizes glass transition temperature (T_g), crystallization temperature (T_c) and melting point (T_{mp}) of the DAEs. The word "sublimation" in the T_{mp} column means that the material was not melted but sublimed. The crystal films were prepared by the following process. The 1- μm -thick amorphous DAE film was deposited on a glass substrate by a conventional vacuum evaporation method. The films were annealed at 40, 60 or 80°C for 48 hours to obtain crystallized films. The word "Yes" in the "Crystallization" column in Table S1 indicates that the amorphous film was converted to the crystal successfully. "No" means that the amorphous film was unchanged or melted. The crystallized films were observed by a crossed-polarizer (Fig. S2). No samples show extraordinarily large crystallites except for the DAE1 film (Fig. 1a).

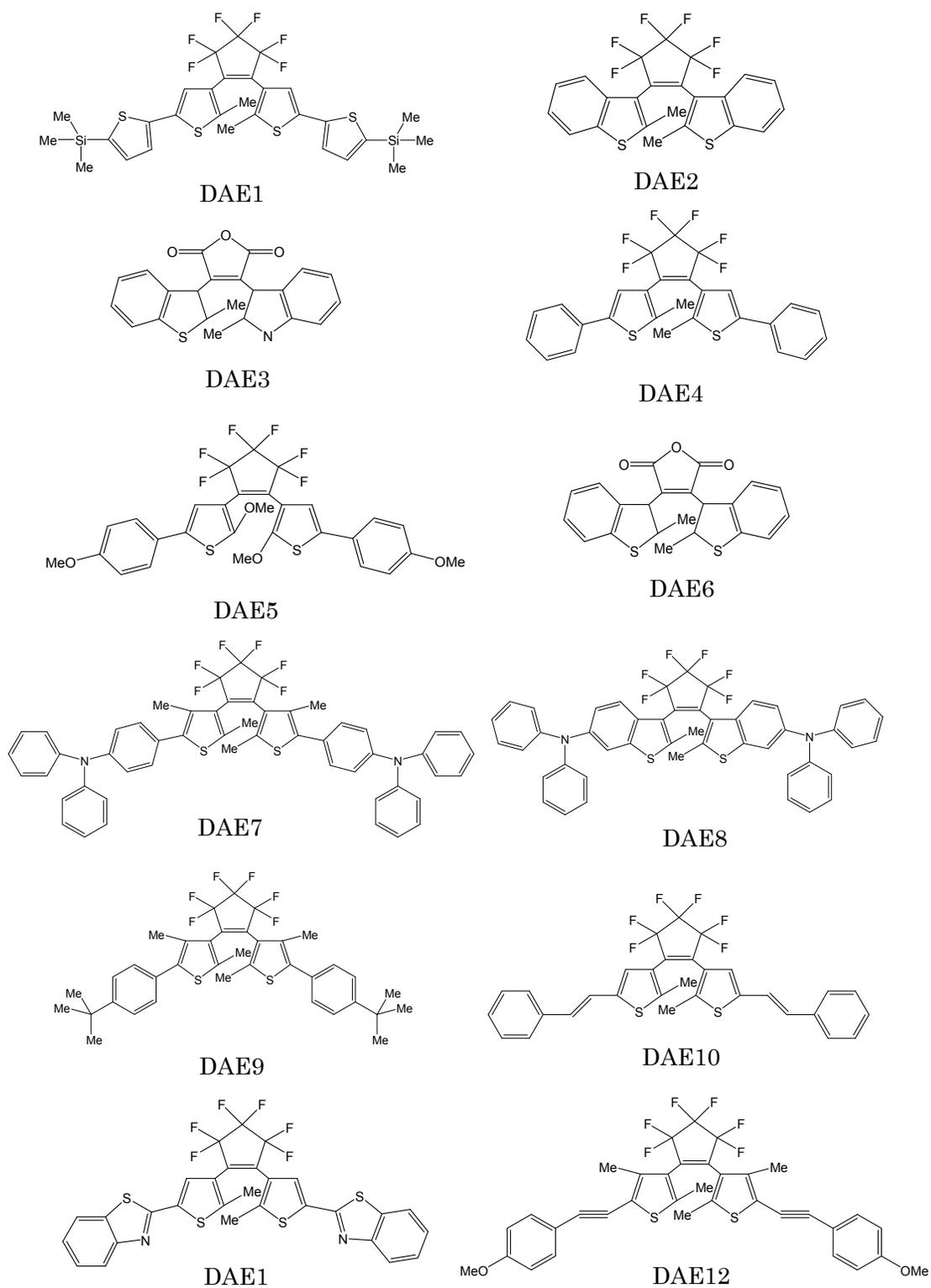
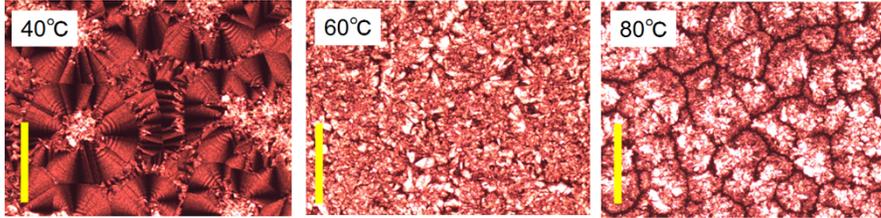


Fig. S1 Molecular structures of DAEs.

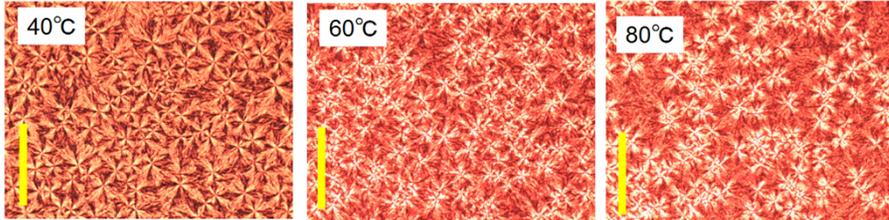
Table S1. Glass transition temperature (T_g), crystallization temperature (T_c) and melting point (T_{mp}) of DAEs.

Sample	T_g / C°	T_c / C°	T_{mp} / C°	Crystallization
DAE1	29	-	158	Yes
DAE2	19	111	156	Yes
DAE3	-	-	sublimation	Yes
DAE4	24	75	139	Yes
DAE5	36	97	157	Yes
DAE6	-	-	sublimation	Yes
DAE7	102	-	105	No
DAE8	106	-	239	No
DAE9	62	-	75	No
DAE10	47	-	145	No
DAE11	76	150	199	Yes
DAE12	47	-	98	No

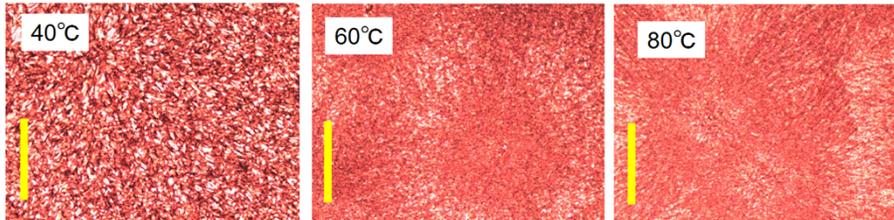
DAE2



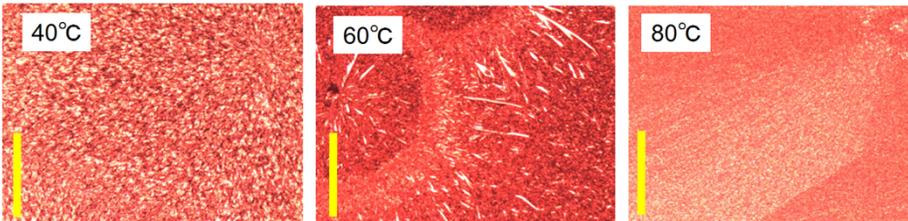
DAE3



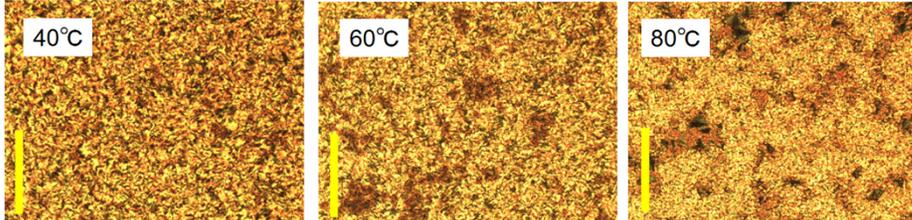
DAE4



DAE5



DAE6



DAE11

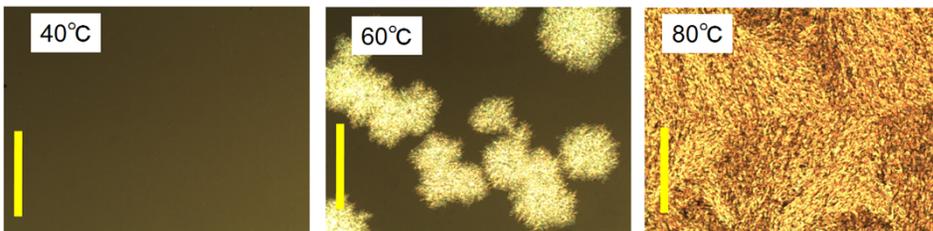


Fig. S2 DAE crystal films observed by crossed-polarizer. Scale bar: 500 μm .

2. Thickness dependence of crystallization of amorphous DAE1 films

Figure S3 shows the thickness dependence of the DAE1 crystal film. The samples were prepared by the following process. Amorphous DAE1 films with various thicknesses were formed on a glass substrate by a vacuum evaporation method. The obtained amorphous DAE1 films were annealed at 60°C for 48 hours to change the amorphous film to the crystal. All samples had extraordinarily large crystallites, but the excessively thick sample (2.5- μm -thick sample) had a wavy surface (upper-left). Samples with less thickness showed relatively flat surfaces.

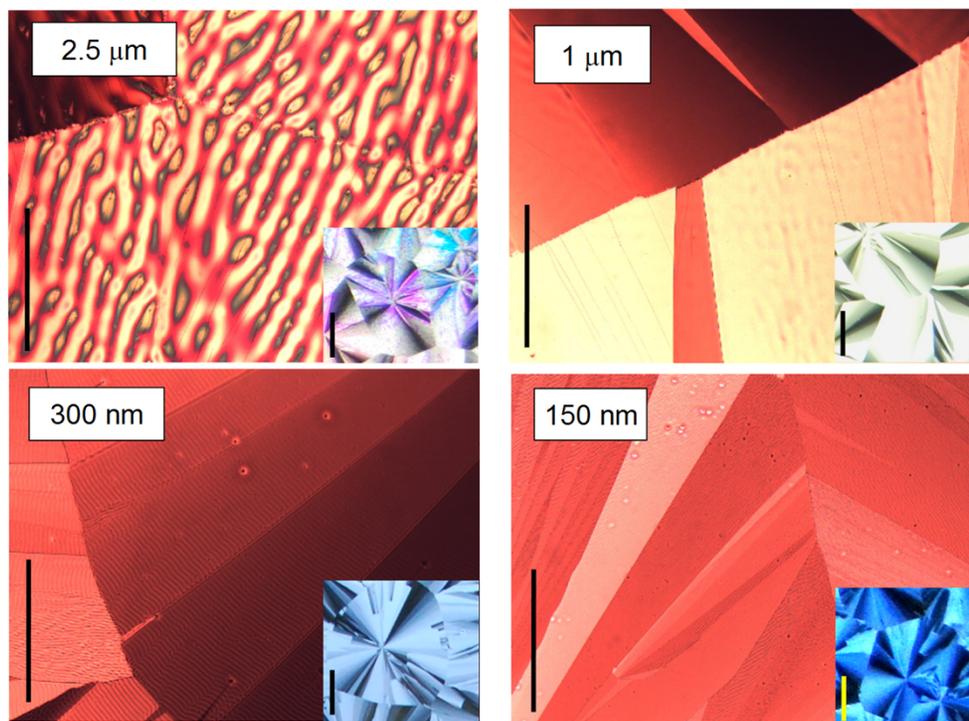


Fig. S3 Thickness dependence of crystal DAE1 film surface. Scale bars in each figure indicate 500 μm and 5 mm (inset).

3. Absorption of DAE1 film

Figure S4a shows the absorption spectrum of DAE1. The colorless state has absorption in the UV region, but the colored state obtained by UV irradiation has both UV and visible regions. In the colored state, the absorbance at 365 nm, which is the wavelength of the UV light for the coloring reaction, is considerably larger than that in the colorless state. Figure S4b shows the calculated transmittance at 365 nm for the colored state. The transmittance exponentially decreases with the film thickness, and the UV light only reaches the 400-nm-depth. This means that UV irradiation isomerizes the colorless crystal layer only near the ITO interface, and the deep part of the film remains in the colorless crystal state.

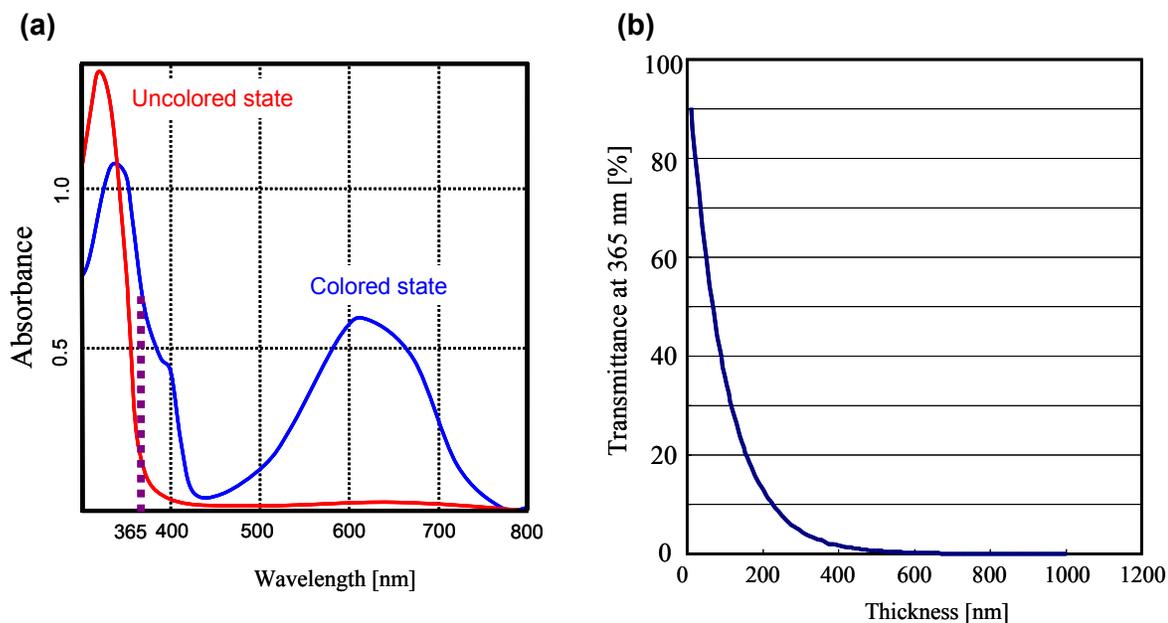


Fig. S4 (a) Absorption spectrum of DAE1 film and (b) calculated transmittance of DAE1 film in colored state

4. Surface roughness of glass and ITO substrates

Figures S5a and S5b show atomic force microscopy (AFM) images of the surfaces on glass and ITO substrates, respectively. Both substrates have a roughness of 5-6 nm, but the glass surface shows better flatness. No difference of the surface flatness, however, was observed on the crystal surfaces growth on the both surfaces: the step and terrace on the DAE1 crystal surface like as shown in Fig. 1c was observed. The size of DAE crystallite was not depending on surface roughness of the ITO electrode.

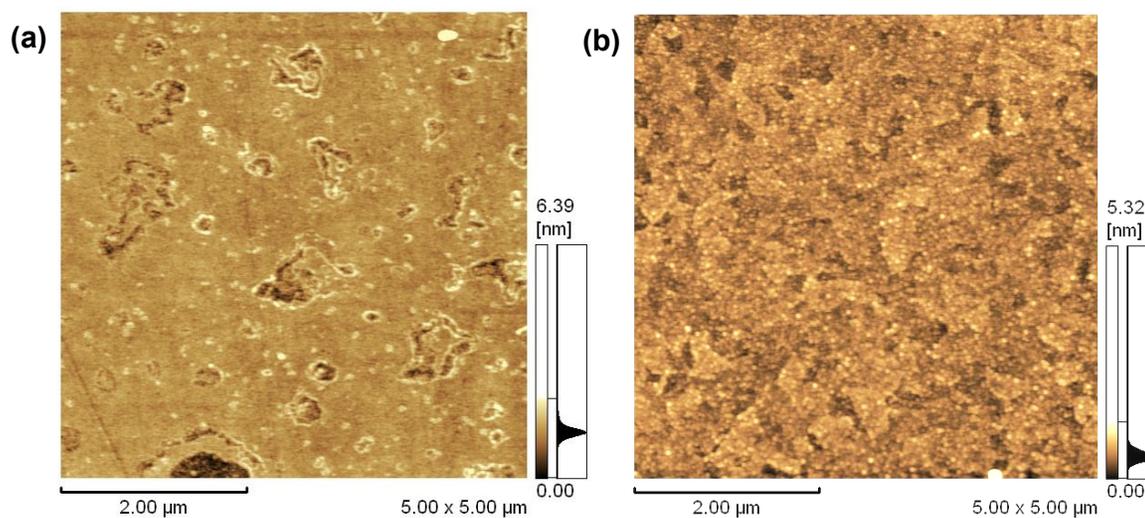


Fig. S5 AFM images of surfaces on (a) glass substrate and (b) ITO substrate