Electronic Supporting Information

Dual-Responsive Benzylidene Indanone Crystals: Mechanical Flexibility Coupled with Reversible Acidochromism

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S1. Synthesis Scheme

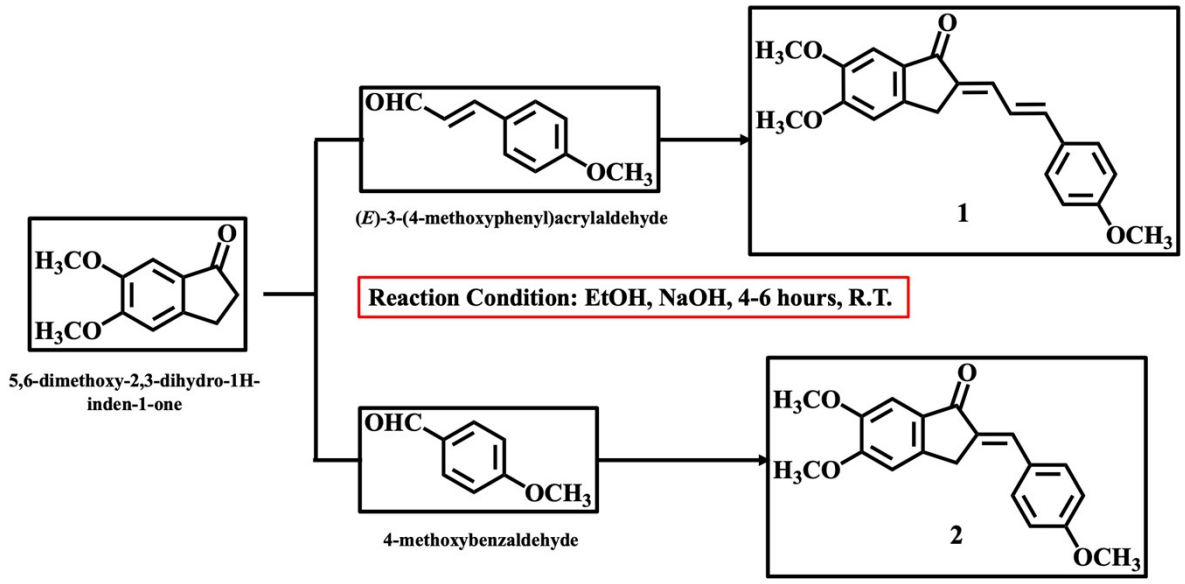


Fig. S1. Synthesis route of compound **1** and compound **2**

Compounds **1** and **2** were synthesized following a previously reported procedure.[1] Specifically, 4,5-dimethoxy-1-indanone (1 mmol) was dissolved in a 100 ml round-bottom flask containing 15 ml of ethanol and a sodium hydroxide solution. Subsequently, trans-4-methoxycinnamaldehyde (1 mmol) and 4-methoxybenzaldehyde (1 mmol) were added to the resulting solution, respectively. The mixture was stirred for 4-6 hours under ambient conditions, leading to the formation of precipitates of compounds **1** and **2** (Scheme S1) with satisfactory yields (Compound **1**: ~85% and Compound **2**: ~90%).

S2. Recrystallisation Table

Table S1. Results from the recrystallisation of Compounds **1** and **2** from one solvent or mixture of solvents.

S. No.	Solvents	Compound 1	Compound 2
1.	Methanol	Powder	Fine Needles
2.	Chloroform	Powder	Powder
3.	Acetone	Powder	Fine Needles
4.	Dichloromethane + Methanol (1:1)	Powder	Powder
5.	Methanol + Chloroform (1:1)	Fine Needles	Fine Needles
6.	Acetonitrile	Powder	Powder
7.	Acetone + Chloroform	Powder	Powder
8.	Acetone + Methanol (1:1)	Powder	Fine Needles
9.	Chloroform + Acetonitrile (1:1)	Fine Needles	Powder

S3. Nuclear Magnetic Resonance (NMR) Analysis.

^{1}H NMR spectra of both Crystal **1** and Crystal **2** were taken in $CDCl_3$ solvent. ^{1}H NMR spectra were collected under standard conditions. Chemical shifts (δ) were reported in parts per million (ppm) and referenced to the residual solvent peak at 7.26 ppm for the $CDCl_3$ solvent peak and 1.60 ppm for the $CDCl_3$ water peak.

Crystal 1: ^{1}H NMR (400 MHz, $CDCl_3$), δ 3.74 (s, 2H, methyl-H), δ 3.84 (s, 3H, methoxy-H), δ 3.94 (s, 3H, methoxy-H), δ 3.99 (s, 3H, methoxy-H), δ 7.02 – 6.82 (m, 5H, aromatic-H and vinylic-H), δ 7.39 – 7.24 (m, 2H, aromatic-H and vinylic-H), δ 7.47 (d, J = 8.7 Hz, 2H, aromatic-H). (Fig. S2).

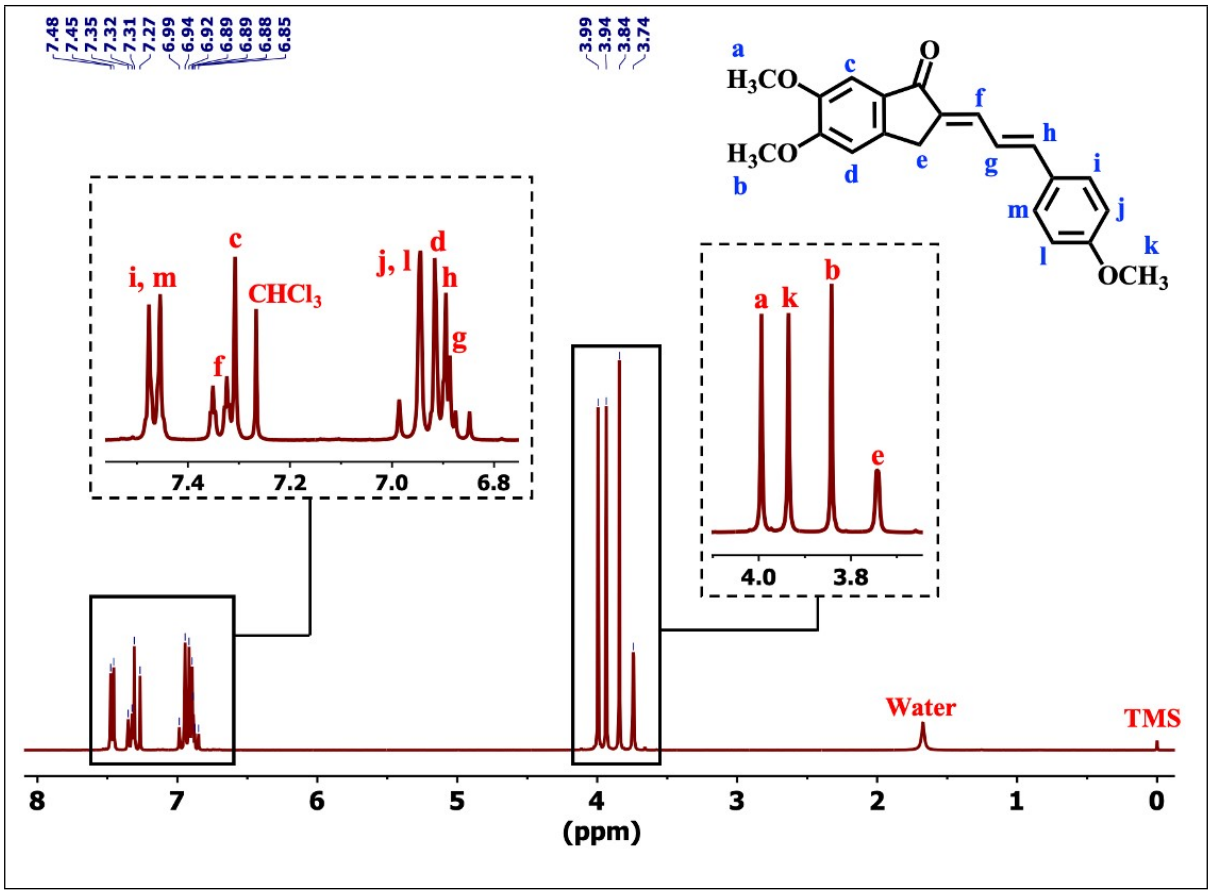


Fig. S2. ^{1}H NMR spectrum of Crystal **1** (solvent: $CDCl_3$, peak at 7.26 ppm).

Crystal 2: ^{1}H NMR (500 MHz, $CDCl_3$), δ 3.89 (s, 3H, methoxy-H), δ 3.95 (s, 2H, methyl-H), δ 3.97 (s, 3H, methoxy-H), δ 4.02 (s, 3H, methoxy-H), δ 6.99 (d, *J* = 8.4 Hz, 3H, aromatic-H), δ 7.36 (s, 1H, aromatic-H), δ 7.58 (s, 1H, vinylic-H), δ 7.64 (d, *J* = 8.7 Hz, 2H, aromatic-H). (Fig. S3).

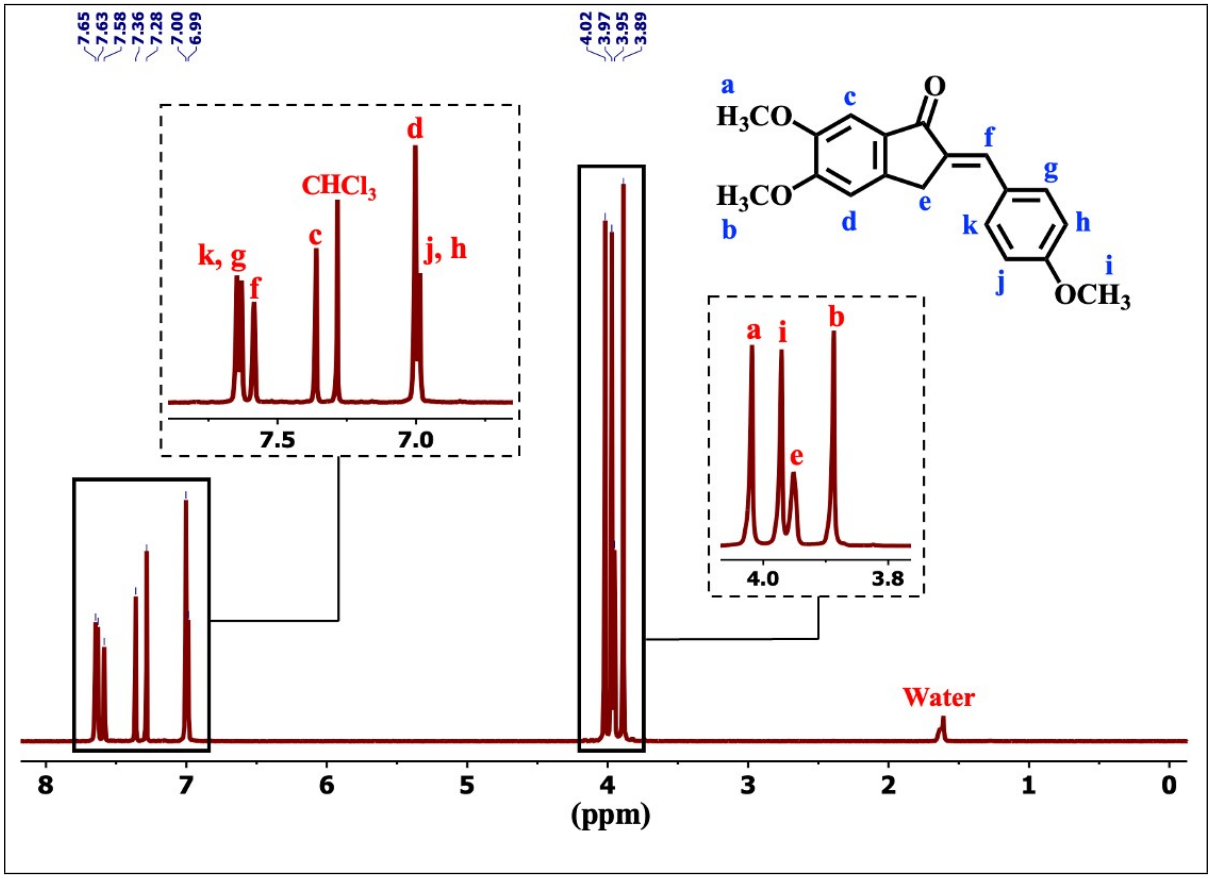


Fig. S3. ^{1}H NMR spectrum of crystal **2** (solvent: $CDCl_3$, peak at 7.26 ppm).

S4. Differential Scanning Calorimetry (DSC) Analysis.

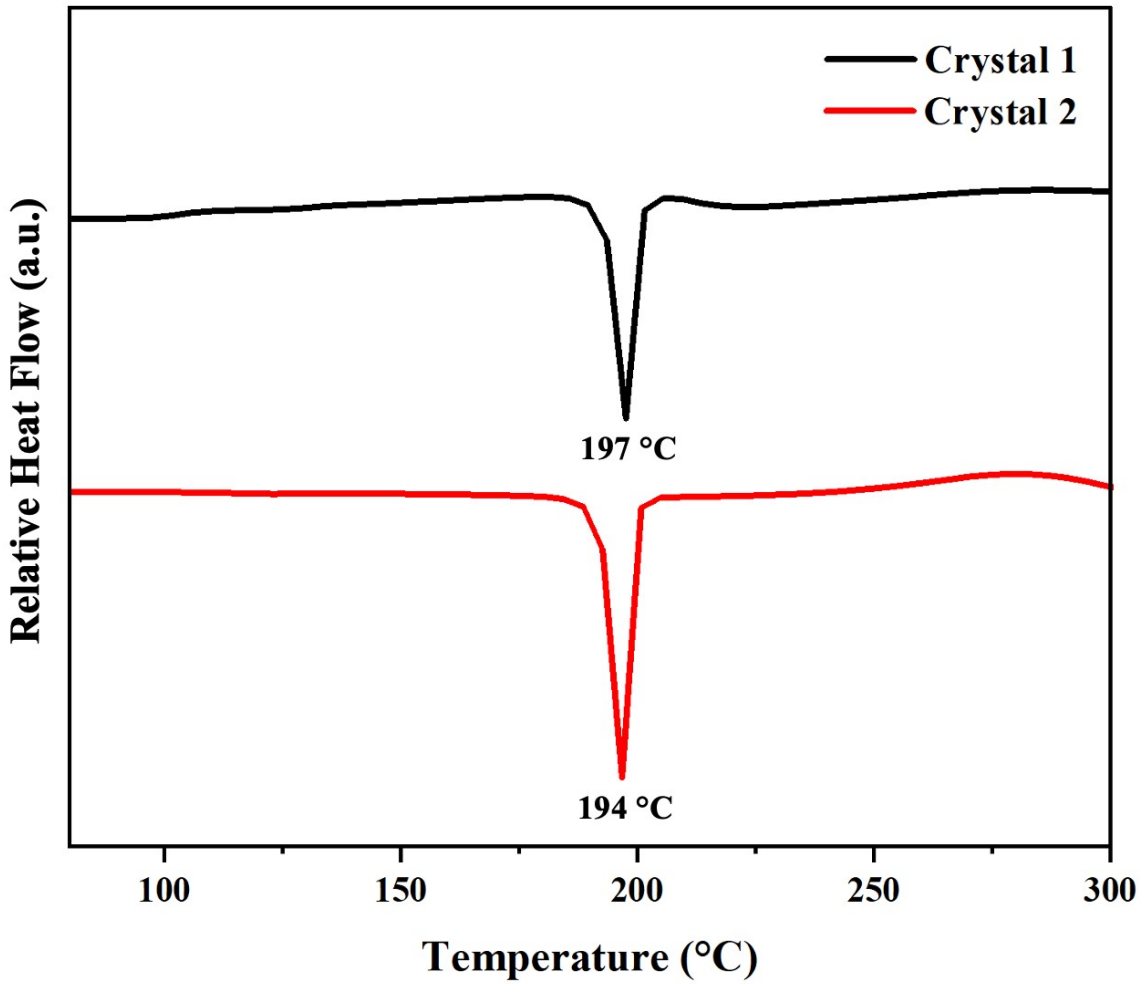


Fig. S4. Differential Scanning Calorimetry (DSC) overlay diagram of crystals **1** and **2**.

S5. Crystallographic Information Table.

Table S2: Crystallographic information table.

Compound	**Crystal 1**	**Crystal 2**
Formula	C_{21} H_{20} O_4	$C_{19}H_{17}O_4$
Molecular Weight	336.37	309.32
T/K	298	298
Crystal System	Monoclinic	Monoclinic
Space Group	$P2_1/c$	$P2_1/c$
a/Å	4.6803(9)	4.8459(8)
b/Å	40.269(7)	35.354(6)
c/Å	9.2487(17)	9.1074(15)
α/°	90	90
β/°	96.433(5)	97.576(5)
γ/°	90	90
Volume/Å^3	1732.1(6)	1546.7(4)
Z	4	4
ρ, Mg.cm^{-3}	1.290	1.328
μ /mm^{-1}	0.089	0.093
Reflections Collected	32841	33608
Independent Reflections	3070	2655
R_{int}	0.1026	0.1492
GOF	1.135	1.061
Final R[I>2σ]	0.0962	0.0595
R_1	0.2232	0.1380
wR_2	0.2463	0.2108
CCDC Number	2492718	2492719

S6. Face Indexing Image.

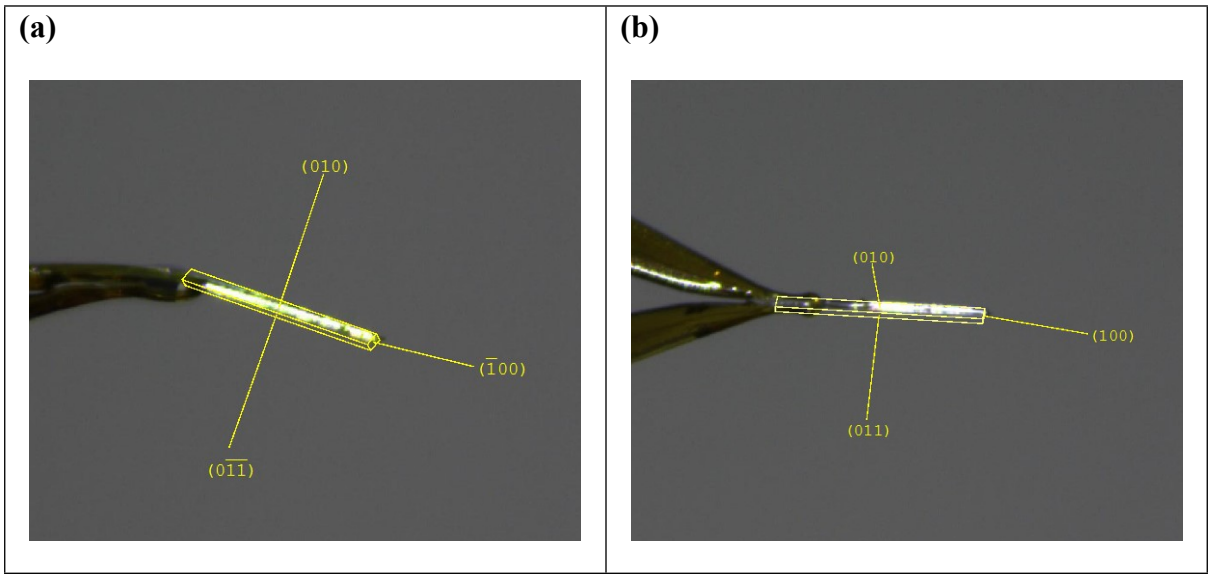


Fig. S5: Face indexing image of **(a)** Crystal **1**, **(b)** Crystal **2**.

S7. Stepwise Bending Images of Crystal 1.

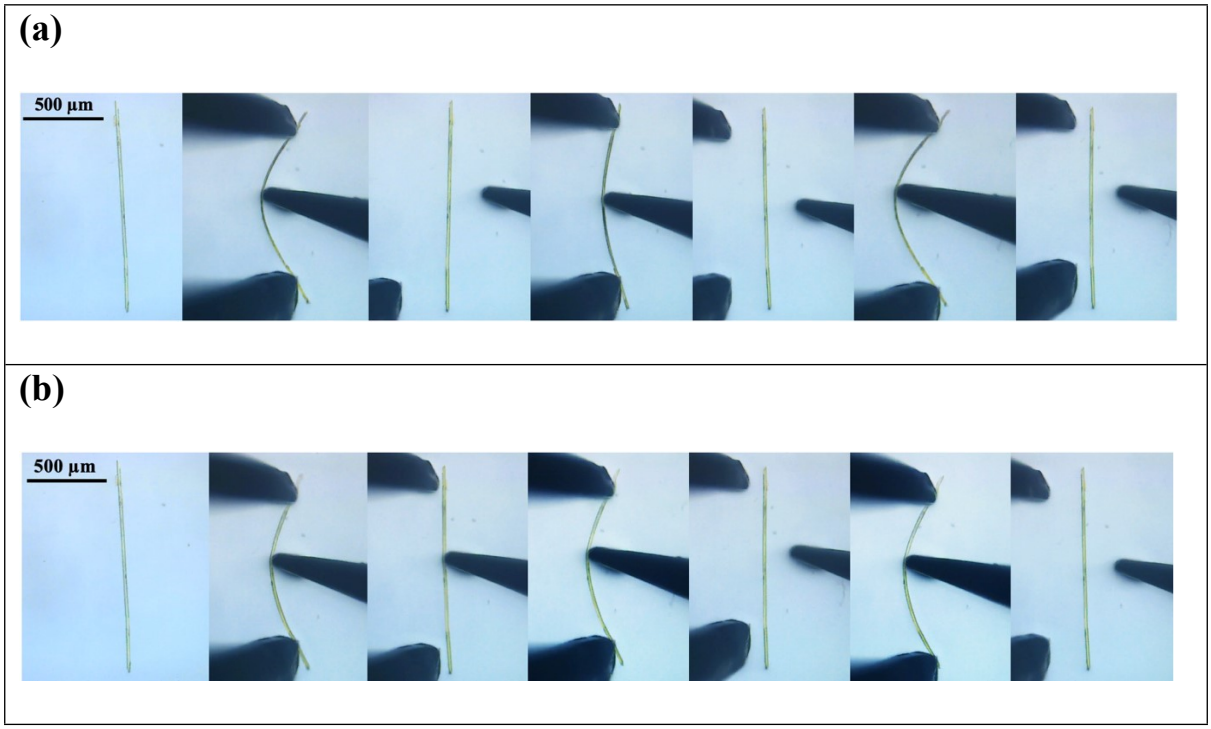


Fig. S6: Stepwise elastic bending images of crystal **1 (a)** (010) face **(b)** (011) face.

S8. Elastic Strain Calculation.

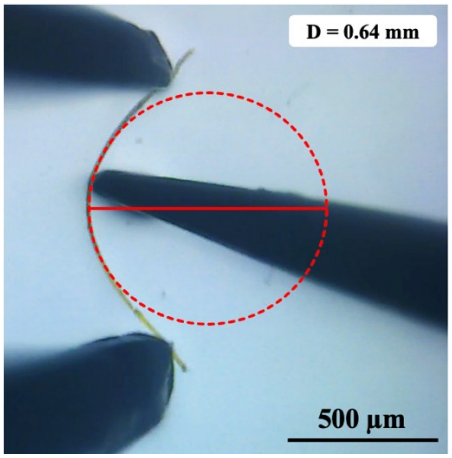
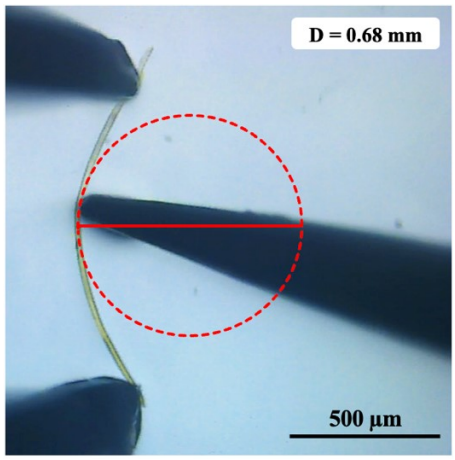
(a)  	**R** = d/2 = 0.32 mm, (where d is the diameter of the semicircle formed and so the radius is d/2) For a beam with a thickness, t, $\varepsilon\ (\%) = \frac{t}{2R} \times 100 = \frac{0.02}{0.64} \times 100 =$ **3.1 %** where 'ε (%)' is the elastic strain of the crystal, 't' is the thickness of the crystal, and 'R' is the radius of the semi-circle formed by bending the crystal.
(b)  	**R** = d/2 = 0.34 mm, (where d is the diameter of the semicircle formed and so the radius is d/2) For a beam with a thickness, t, $\varepsilon\ (\%) = \frac{t}{2R} \times 100 = \frac{0.02}{0.68} \times 100 =$ **2.9 %** where 'ε (%)' is the elastic strain of the crystal, 't' is the thickness of the crystal, and 'R' is the radius of the semi-circle formed by bending the crystal.

Fig. S7: Elastic strain calculation of Crystal **1** from (010) face (a) and (011) face (b).

S9. Solution State Absorption and Emission Studies.

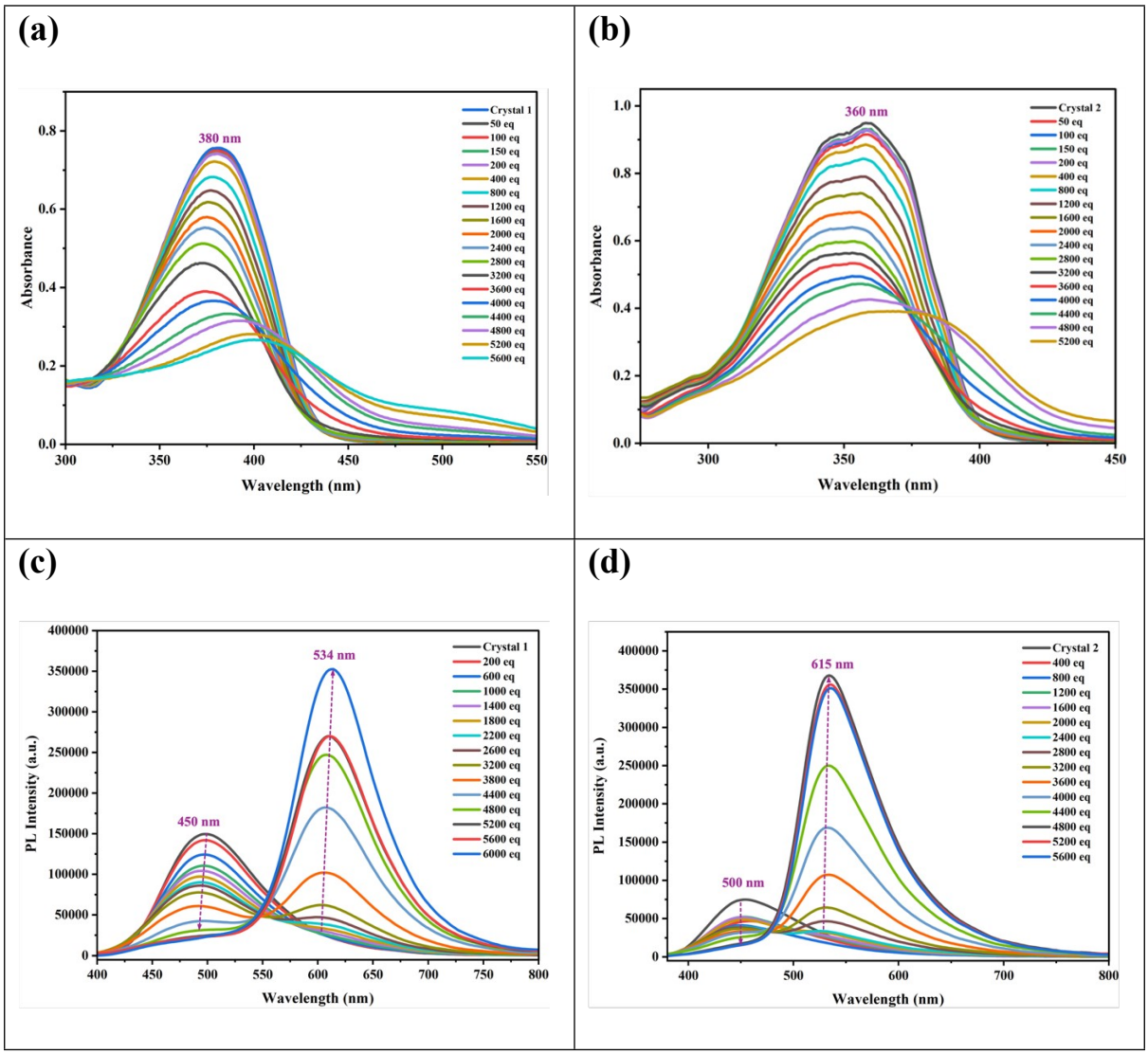


Fig. S8. (a) and **(b)** Solution state absorption, and **(c)** and **(d)** emission spectra (6×10^{-6} M) of crystals **1** and **2** as a function of TFA concentration in DMSO.

S10. FTIR Spectra for Acidochromism.

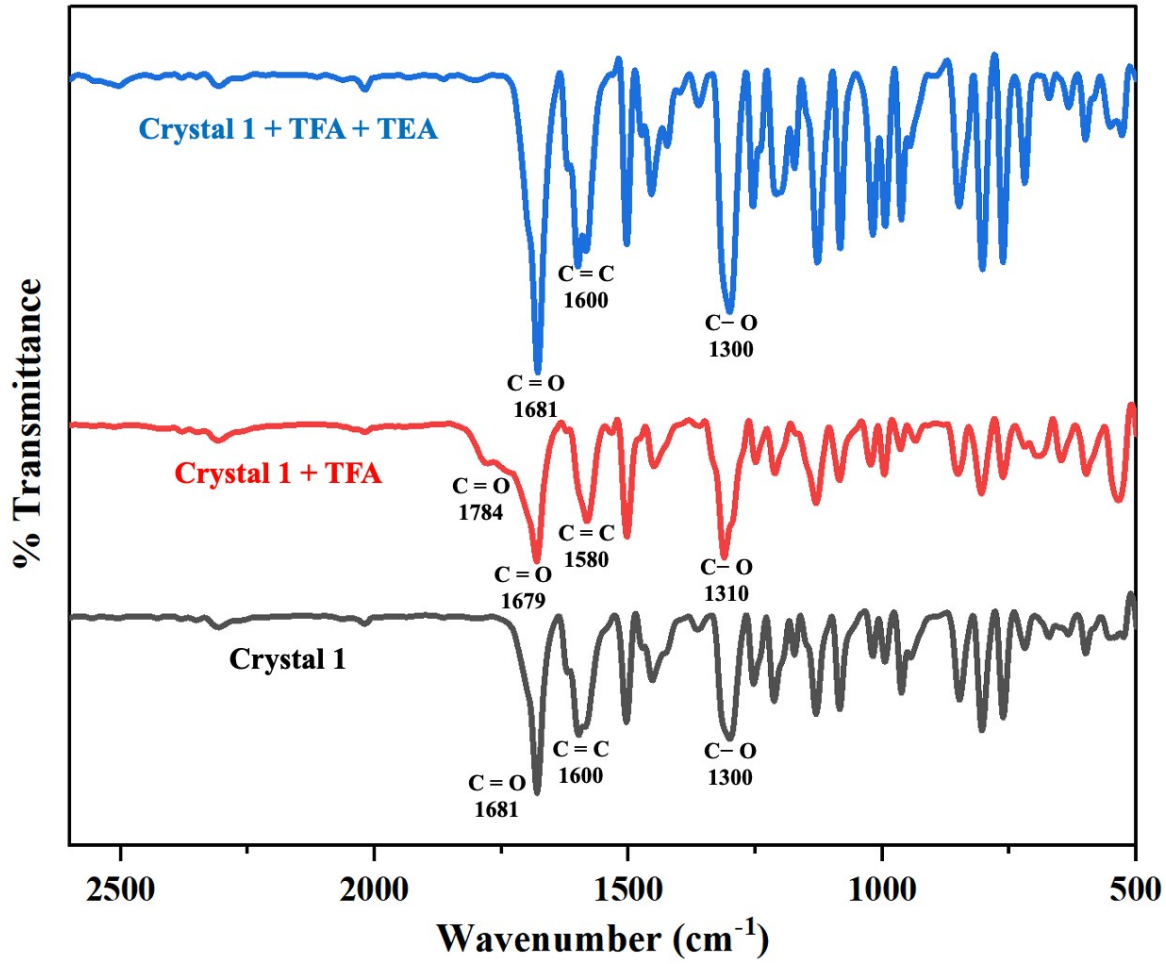


Fig. S9: FTIR overlay spectra of crystals **1** depicting spectra of pristine sample, treated with TFA and TEA.

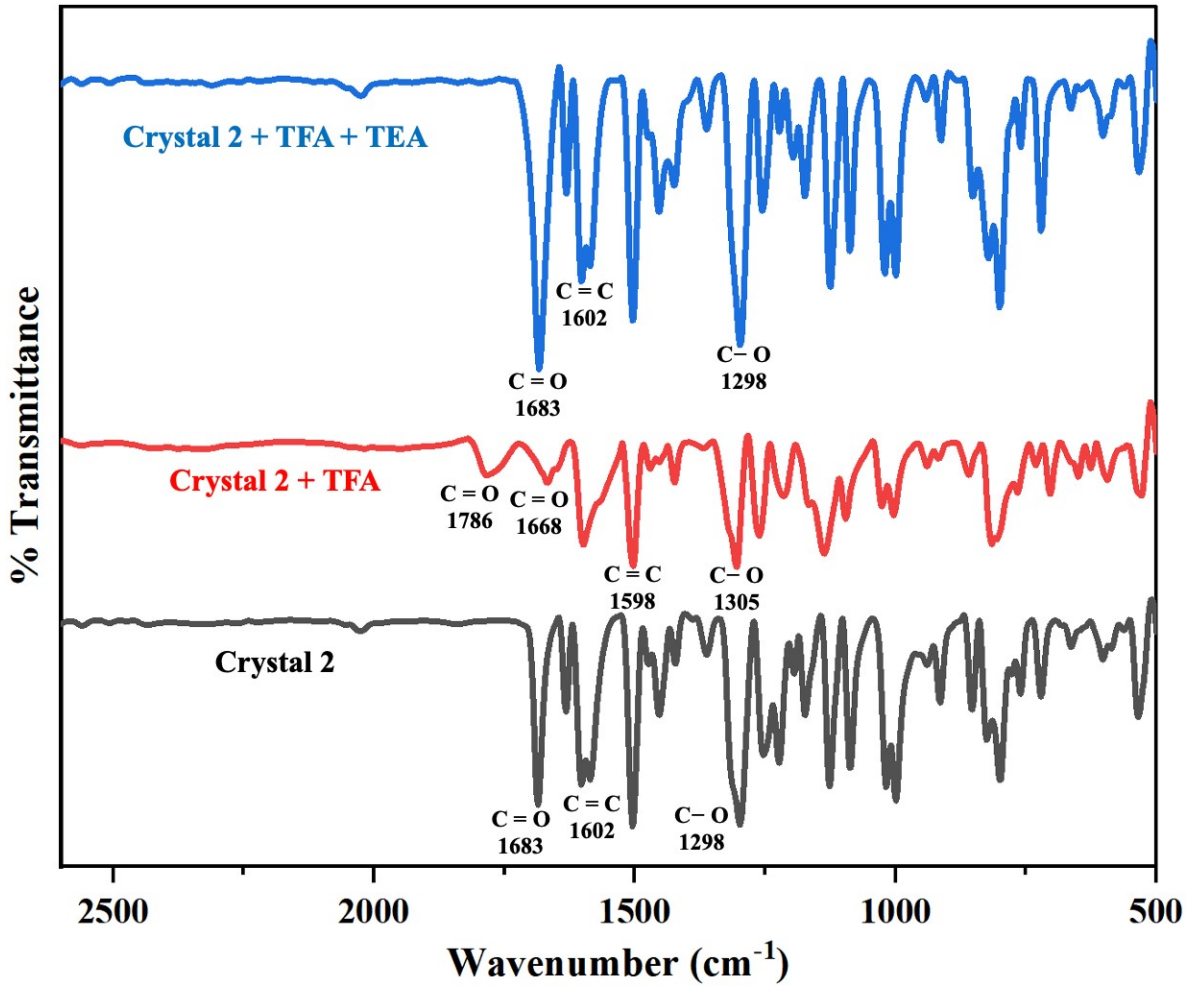


Fig. S10: FTIR overlay spectra of crystals **2** depicting spectra of pristine sample, treated with TFA and TEA.

S11. After Acid Fumigation Elastic Strain calculation.

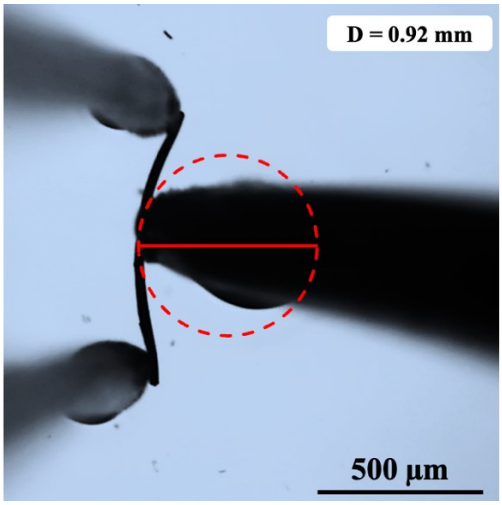
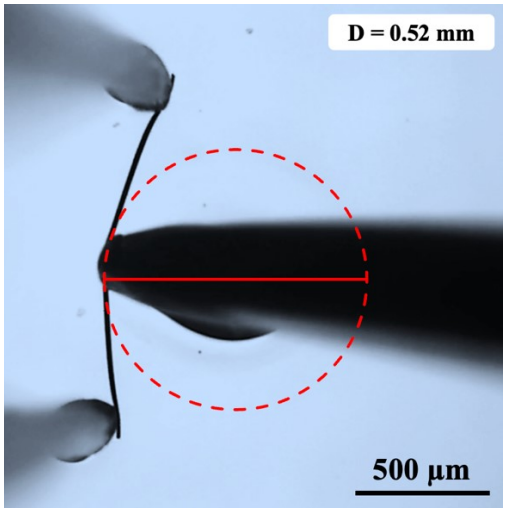
(a)  	**R** = d/2 = 0.46 mm, (where d is the diameter of the semicircle formed and so the radius is d/2) For a beam with a thickness, t, $\varepsilon\ (\%) = \frac{t}{2R} \times 100 = \frac{0.016}{0.92} \times 100 =$ **1.7 %** where 'ε (%)' is the elastic strain of the crystal, 't' is the thickness of the crystal, and 'R' is the radius of the semi-circle formed by bending the crystal.
(b)  	**R** = d/2 = 0.26 mm, (where d is the diameter of the semicircle formed and so the radius is d/2) For a beam with a thickness, t, $\varepsilon\ (\%) = \frac{t}{2R} \times 100 = \frac{0.012}{0.52} \times 100 =$ **2.3 %** where 'ε (%)' is the elastic strain of the crystal, 't' is the thickness of the crystal, and 'R' is the radius of the semi-circle formed by bending the crystal.

Fig. S11. Elastic strain calculation of Crystal **1** + TFA **(a)** (011) face **(b)** (010) face.

S12. Overall Elastic Strain Calculation Table.

Table S3: Overall Elastic Strain Calculation Table

S. No	Name	Elastic Strain Values 'ε (%)'	
		(010) face	(011) face
1.	Crystal **1**	3.1	2.9
2.	Crystal **1** + TFA	2.3	1.7

S13. References:

1. R. Ahamed, A. Bhowmik, M. K. Mishra and S. Ghosh, *Cryst. Growth Des.*, 2025, **25**, 4481 – 4493.