

Supplementary Materials for

High-Speed Steerable Polar Crystal Motors Powered by Photoinduced Inverse Piezoelectricity

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I. General information

Materials

Unless otherwise mentioned, all reagents and solvents were purchased from commercial sources and used as received without further purification.

Synthetic characterization

NMR spectra were recorded on an Agilent 400-MR DD2 spectrometer. The ^1H NMR (400 MHz) chemical shifts were measured relative to CDCl_3 as the internal reference (CDCl_3 : $\delta = 7.26$ ppm). High-resolution mass spectra (HRMS) were obtained with a Shimadzu LCMS-ITTOF (ESI).

Spectral data characterization

The UV-vis diffuse reflectance spectrum of the crystals was recorded using a Shimadzu UV-2600 spectrophotometer with BaSO_4 as a standard (100% reflectance). Lifetime measurements of the **H_z-F_G** and **H_z-F_Y** crystals were performed using a Quantaaurus-Tau fluorescence lifetime spectrometer. Quantum yield measurements of the **H_z-F_G** and **H_z-F_Y** crystals were conducted using a Quantaaurus-QY fluorescence quantum yield spectrometer. Emission and second-harmonic generation (SHG) measurements were carried out on a home-built inverted fluorescence microscope system. For these measurements, excitation pulses at 400 nm were generated from the second harmonic of the fundamental output of a regenerative amplifier (Spectra Physics, 800 nm, 150 fs, 1 kHz), which was seeded by a mode-locked Ti:sapphire laser (Mai Tai, Spectra Physics, 800 nm, 150 fs, 80 MHz). The laser beam was focused to a spot diameter of 40 μm through an objective lens (Nikon CFLU Plan, 50 $\times$, N.A. = 0.8). The excitation energy was varied using a neutral density filter.

Crystallographic characterization

Single-crystal X-ray diffraction data were collected on an Agilent Technologies Gemini diffractometer. Transmission electron microscopy (TEM) images and selected area electron diffraction (SAED) patterns were obtained using a JEOL JEM-F200 microscope operated at an accelerating voltage of 200 kV. All samples were prepared by drop-casting the micro-ribbon suspensions onto a copper grid coated with a thin carbon support film. Powder X-ray diffraction (PXRD) data were collected using a Bruker D2 Phaser diffractometer with $\text{Cu-K}\alpha$ radiation ($\lambda = 1.5416$ Å) at 30 kV and 10 mA.

Laser-driven crystal motion testing

The optically driven motion of **H_z-F_G** and **H_z-F_Y** molecular crystals was investigated using a programmable laser scanning microscope system, with the motion recorded by a camera. This system primarily consists of a stable-output laser source (VIS-MLD, 405 nm) and a galvanometer scanning system (LightScan), enabling arbitrary control of the laser scanning trajectory on the substrate. A CCD camera (Dhyana 400BSI V3) and a high-numerical-aperture 20× objective (Olympus, N.A. = 0.4) were used for imaging, and the captured images were subsequently analyzed for crystal motion. Prior to measurements, the laser spot size and the conversion factor between pixel and physical dimensions were calibrated using a standard target.

The molecular crystals were transferred onto OTS-functionalized transparent glass substrates via a poor-solvent transfer method for motion testing. Before modification, pristine glass slides were immersed in piranha solution (H₂SO₄/H₂O₂, 3:1, v/v) for 1.5 h, followed by thorough rinsing with acetone and toluene. The slides were then immersed in a toluene/OTS mixture (100:1, v/v) for 3 h, rinsed repeatedly with toluene and acetone, and finally dried to obtain the OTS-modified substrates.

Bulk photovoltaic effect (BPVE) device testing

Electrical contacts were made using carbon-paste electrodes. The current-voltage (*I*-*V*) characteristics of the **H_z-F_G** and **H_z-F_Y** crystals were measured between two carbon-paste electrodes at room temperature using a Keithley 2400 SourceMeter. To determine the effective contact area, the cross-sectional profiles of the crystal-electrode interfaces were measured using a Bruker DektakXT stylus profiler; the raw *I*-*V* data were subsequently converted to current density-voltage (*J*-*V*) curves. Illumination experiments were performed using a Newport Sol3A Class AAA solar simulator, with the irradiance set to 100 mW cm⁻² (equivalent to 1 sun).

Piezoresponse force microscopy (PFM) characterization

PFM measurements were conducted using a commercial atomic force microscope system (Bruker, Dimension Icon) on **H_z-F_G** and **H_z-F_Y** single crystals. Conductive platinum-iridium coated silicon probes (SCM-PIT-V2) with a spring constant of approximately 3 N m⁻¹ were utilized to probe the piezoelectric properties of the crystals. To determine the local effective piezoelectric coefficients, an AC drive voltage ranging from 0 V to 5 V was applied. Data points collected between 1 V and 5 V were selected for linear slope fitting to exclude potential electrostatic contributions or nonlinear responses under low voltages; local effective piezoelectric coefficients were calculated from the slopes of the resulting amplitude-excitation curves. All acquired data were analyzed using NanoScope Analysis 3.0 software, and measurements were performed at three independent regions for each sample.

Switching spectroscopy PFM (SS-PFM) was further adopted to characterize local ferroelectric domain switching and polarization reversal behaviors. Cyclic DC bias voltages ranging from -10 V to 10 V were applied to **H_z-F_G** and **H_z-F_Y** crystals, and characteristic piezoresponse amplitude butterfly loops together with phase hysteresis curves were recorded.

Tensile Strength Tests

Tensile mechanical measurements were performed on a CARE IBTC-300SL in situ tensile testing system equipped with high-precision displacement and load sensors. The instrument has a displacement resolution of 0.1 μm and a load resolution of 1 mN. All tests were conducted under displacement control at a constant rate of 5 $\mu\text{m}/\text{min}$. The initial spacing of the fixed crystal sample before stretching was 4 mm. They were mounted using dedicated tensile grips, and a small amount of adhesive was applied to minimize relative slippage during deformation. The system uses a symmetric center-loading configuration that keeps the specimen at a fixed position during loading, enabling stable mechanical measurements compatible with in situ characterization.

During the tests, load (F) and displacement (d) data were recorded in real time. The stress, strain and elastic modulus were calculated by the Equations¹:

$$\text{Elastic Modulus} = \frac{\text{Stress}}{\text{Strain}}$$

$$\text{Stress (MPa)} = \frac{F}{A} = \frac{\text{Applied force}}{\text{Cross-section area}}$$

$$\text{Strain} = \frac{\Delta L}{L} = \frac{\text{Change in length}}{\text{Initial length}}$$

where A is the initial cross-sectional area, ΔL is the displacement measured by the system, and L is the original length. The elastic modulus (E) was then obtained as the slope of the linear region of the resulting stress-strain curve.

Raman measurements

Prior to use, the sample stage was rinsed repeatedly with ethanol (2-3 times) and dried under a nitrogen stream. Molecular crystals of **H₂-F₆**, **H₂-F₇** and **H₂-Me** were then transferred onto a clean glass slide. Raman measurements were performed using a RamBo 620 Raman spectrometer (Ntegra RamBo, NT-MDT, Netherlands) equipped with a 100 $\times$ objective lens (Mitutoyo, NA = 0.7) and a 633 nm laser (spot size $\sim$ 1 μm).

The calculation principle and algorithm for the analysis of the crystal elongation before and after irradiation

Computational Principle: Crystal deformation was quantified from optical microscopy images using an image-based geometric measurement workflow. Images of the same crystal were acquired before and after laser irradiation, denoted as l_0 and l_1 , respectively. After image enhancement, object segmentation, sub-pixel edge localization, and maximum Feret diameter measurement, the characteristic lengths of the crystal along its principal axis were obtained as L_0 and L_1 . The absolute elongation of the crystal was defined as:

$$\Delta L = L_1 - L_0$$

The physical lengths were converted from pixel measurements using a calibration factor s , expressed in $\mu\text{m}/\text{pixel}$. If the measured pixel lengths before and after laser irradiation are l_0 and l_1 respectively, the calibrated lengths and elongation are given by:

$$L_0 = l_0 s, L_1 = l_1 s, \Delta L = (l_1 - l_0) s$$

The accuracy of this approach depends primarily on robust boundary localization and the use of a reproducible geometric descriptor for crystal length. Local contrast was first enhanced by contrast limited adaptive histogram equalization (CLAHE), which improves the gray-level separation

between the crystal and the background. The crystal region was then extracted using either Otsu automatic thresholding or a user-defined threshold. Connected components were filtered according to area, contact with the image border, aspect ratio, and compactness to suppress background artifacts and retain the target crystal region.

For sub-pixel edge localization, the external contour of the binary crystal mask was first extracted. At each contour point, the local tangent was estimated from neighboring points, and the corresponding normal direction was used for gray-level sampling across the boundary. The threshold-crossing position along each normal profile was refined by linear interpolation, yielding a sub-pixel contour. A convex hull was then constructed from the refined contour points, and the maximum pairwise Euclidean distance on the hull was taken as the maximum Feret diameter. This value was used as the characteristic crystal length because it represents the longest projected dimension of the segmented object.

In addition, global translational drift between the two images was estimated by phase correlation. Both images were multiplied by a Hann window and transformed using the two-dimensional fast Fourier transform. The normalized cross-power spectrum was computed, and the inverse Fourier transform yielded a correlation surface whose peak indicates the global displacement. The peak position was further refined by parabolic interpolation to obtain sub-pixel drift components. These drift values were used as auxiliary diagnostic information for assessing field-of-view shifts between the two acquisitions.

Algorithmic Workflow: Input: Pre-irradiation image l_0 , post-irradiation image l_1 , pixel calibration factor s ($\mu\text{m}/\text{pixel}$), optional regions of interest R_0 and R_1 , and segmentation parameters. Output: Pre-irradiation length L_0 , post-irradiation length L_1 , elongation ΔL , sub-pixel contours, Feret endpoints, and image drift (dx , dy).

1. Convert l_0 and l_1 to grayscale images when required.
2. Enhance local contrast using CLAHE to improve the separability of the crystal from the background.
3. If regions of interest are provided, restrict subsequent processing to R_0 and R_1 .
4. Segment each enhanced image using Otsu automatic thresholding or a user-defined threshold. The threshold polarity is selected according to whether the crystal is darker or brighter than the background.
5. Filter connected components by area, image-border contact, aspect ratio, and compactness, and retain the largest valid component as the crystal mask.
6. Apply morphological closing to repair small discontinuities in the segmented crystal region.
7. Extract the external contour of the crystal mask.
8. For each contour point, estimate the local tangent from neighboring contour points and define the corresponding normal direction.
9. Sample gray-level intensities along the normal direction within a local window around the contour point.
10. Identify the threshold-crossing position along the normal profile and refine the contour position to sub-pixel precision by linear interpolation.
11. Construct the convex hull of the refined contour point set.
12. Search the convex hull for the point pair with the maximum Euclidean distance; define this distance as the maximum Feret diameter l and record the two endpoints.
13. Repeat steps 7-12 for both l_0 and l_1 to obtain the pixel lengths l_0 and l_1 .
14. Convert pixel lengths into physical lengths using the calibration factor:

$$L_0 = l_0 s, L_1 = l_1 s$$

15. Calculate elongation:

$$\Delta L = L_1 - L_0$$

16. Estimate global image drift using phase correlation. Apply Hann windowing, compute the Fourier transforms of I_0 and I_1 , calculate the normalized cross-power spectrum, and determine (dx, dy) from the correlation peak with sub-pixel parabolic interpolation.

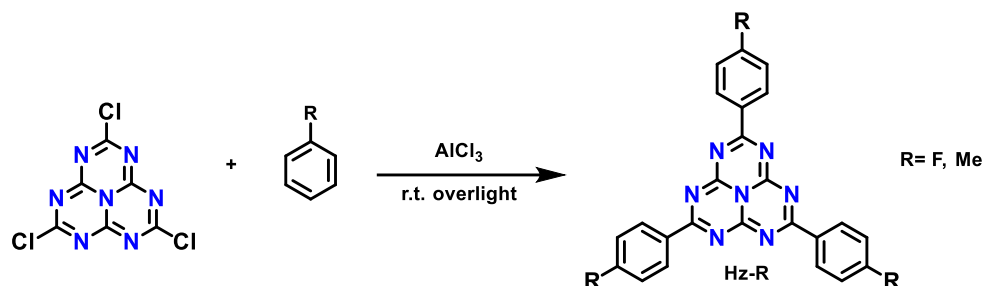
17. Export the measurement results, including the original images, sub-pixel contours, Feret diameter endpoints, calibrated lengths, elongation, pixel size, and drift values.

To reduce the deviation caused by background artifacts, the same segmentation parameters were used to analyze the images of the target crystal before and after irradiation.

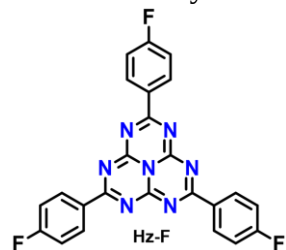
Theoretical calculations

Dipole moments for **H₂-F** and **H₂-Me** in vacuum and crystals were calculated at the ω B97XD/def2-TZVPD level by using the Gaussian 16 program package.² First-principles calculations were performed using the Vienna ab initio Simulation Package (VASP)³⁻⁴ based on density functional theory (DFT) with the projector augmented wave (PAW) method.⁵ The generalized gradient approximation (GGA-PBE) functional of Perdew-Burke-Ernzerhof (PBE)⁶⁻⁷ was utilized to define the exchange-correlation interaction. Elastic tensors and piezoelectric coefficients were calculated using density functional perturbation theory and finite-difference methods.⁸⁻⁹ The converged elastic constants were further processed to derive bulk modulus B , shear modulus G , Young's modulus E , and Poisson's ratio ν using the Voigt-Reuss-Hill averaging scheme. Intermolecular interaction energies for **H₂-F** and **H₂-Me** were calculated using Crystal Explorer 17.5.¹⁰

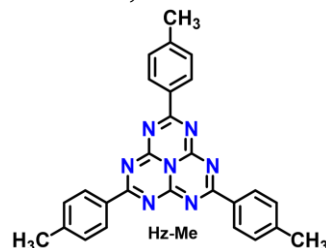
II. Synthesis



Scheme S1. Synthesis routes of **Hz-R**. (R= F, Me)



Synthesis of Hz-F. At 0 °C, anhydrous AlCl_3 (3.8 g, 29.2 mmol) was suspended in fluorobenzene (20 mL). After stirring for 10 minutes, 2,5,8-trichloroheptazine (2 g, 7.3 mmol) was added, turning the mixture deep red. The reaction was stirred vigorously at 0 °C for 30 minutes and then allowed to warm to room temperature and stir overnight. At the end of the reaction, the mixture had turned black. The reaction was quenched by pouring onto a large amount of ice water, and a yellow product was observed to form.¹¹ The product was then collected by filtration and washed three times with acetone to give pure **Hz-F**. (Yield: 1.5 g, 46%) ^1H NMR (400 MHz CDCl_3) δ [ppm] = 7.20 (m, 6H), 8.61 (d, J = 8.0 Hz, 6H). HRMS (ESI⁺): calcd. for $\text{C}_{24}\text{H}_{12}\text{F}_3\text{N}_7\text{Na}^+$: $[\text{M}+\text{Na}]^+$, 478.0999, found: 478.0997.



Synthesis of Hz-Me. The synthetic procedure is totally same with that of **Hz-F** except that toluene was used in place of fluorobenzene. Yield: 1.7 g, 53%. ^1H NMR (400 MHz CDCl_3) δ [ppm] = 2.46 (s, 9H), 7.32 (d, J = 8.0 Hz, 6H), 8.47 (d, J = 8.0 Hz, 6H). HRMS (ESI⁺): calcd. for $\text{C}_{27}\text{H}_{22}\text{N}_7$: $[\text{M}+\text{H}]^+$, 444.1932, found: 444.1935.

III. Crystal growth and characterization

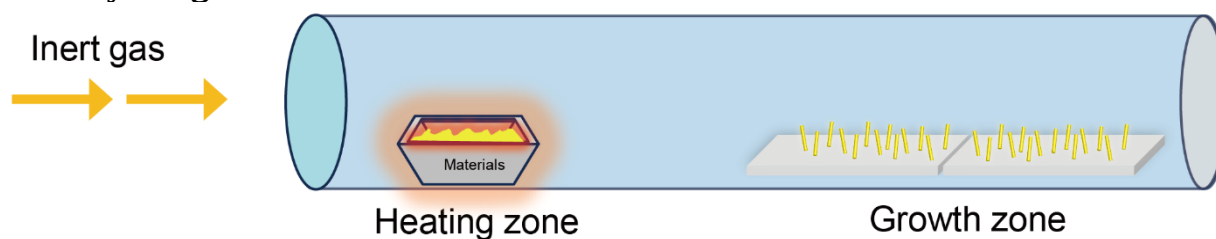


Figure S1. The schematic of the physical vapor transport (PVT) process to prepare crystals.

Table S1. PVT crystal growth conditions.

Materials	Inert gas (mL/min)	Heating zone (°C)	Growth zone (°C)	Heating time (min)
H _z -F _G	30	330	r.t.	60
H _z -F _Y	50	370	r.t.	120
H _z -Me	50	350	r.t.	80

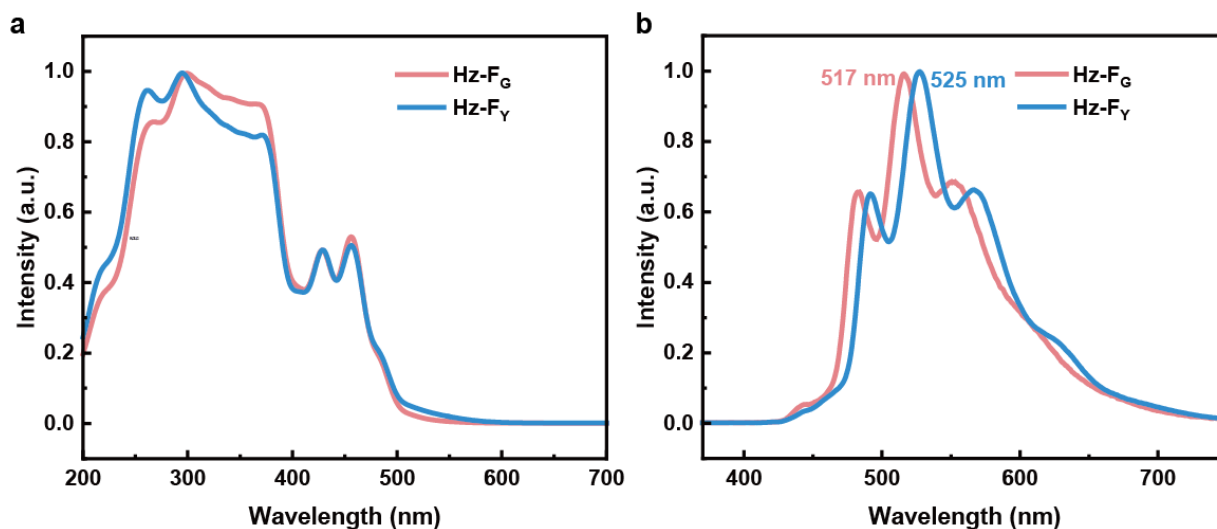


Figure S2. The (a) UV-vis absorption and (b) emission of **H_z-F_G** and **H_z-F_Y** crystals.

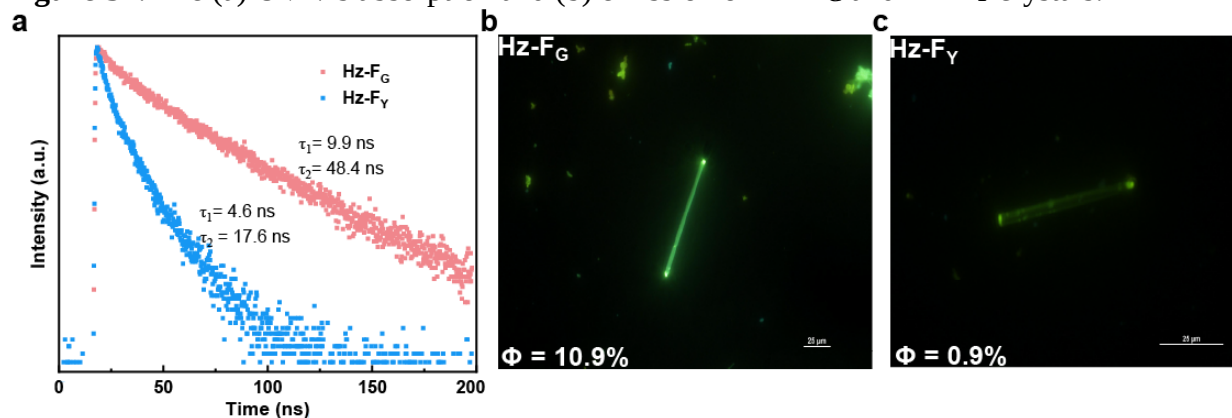


Figure S3. (a) Time-resolved photoluminescence decay curves of **H_z-F_G** and **H_z-F_Y** crystals fitted with a biexponential decay model. (b) Fluorescence microscopy image of a single **H_z-F_G** crystal and (c) a single **H_z-F_Y** crystal, with the measured photoluminescence quantum yields (Φ).

Table S2. The crystal sizes and linear motion velocities within 1 min of two crystals (five **Hz-F_G** and **Hz-F_Y** crystals with similar sizes).

	Sample	Crystal length [μm]	Crystal width [μm]	Speed [μm/s]	Average speed [μm/s]
Hz-F_G	Crystal 1	236.56	8.86	0.77	0.85 ± 0.05
	Crystal 2	255.72	10.14	0.89	
	Crystal 3	248.45	13.60	0.89	
	Crystal 4	193.29	7.04	0.87	
	Crystal 5	260.63	10.78	0.84	
Hz-F_Y	Crystal 1	202.97	6.40	0.91	0.98 ± 0.08
	Crystal 2	224.71	6.40	1.00	
	Crystal 3	242.58	13.06	0.91	
	Crystal 4	226.82	12.83	0.97	
	Crystal 5	262.55	8.83	1.12	

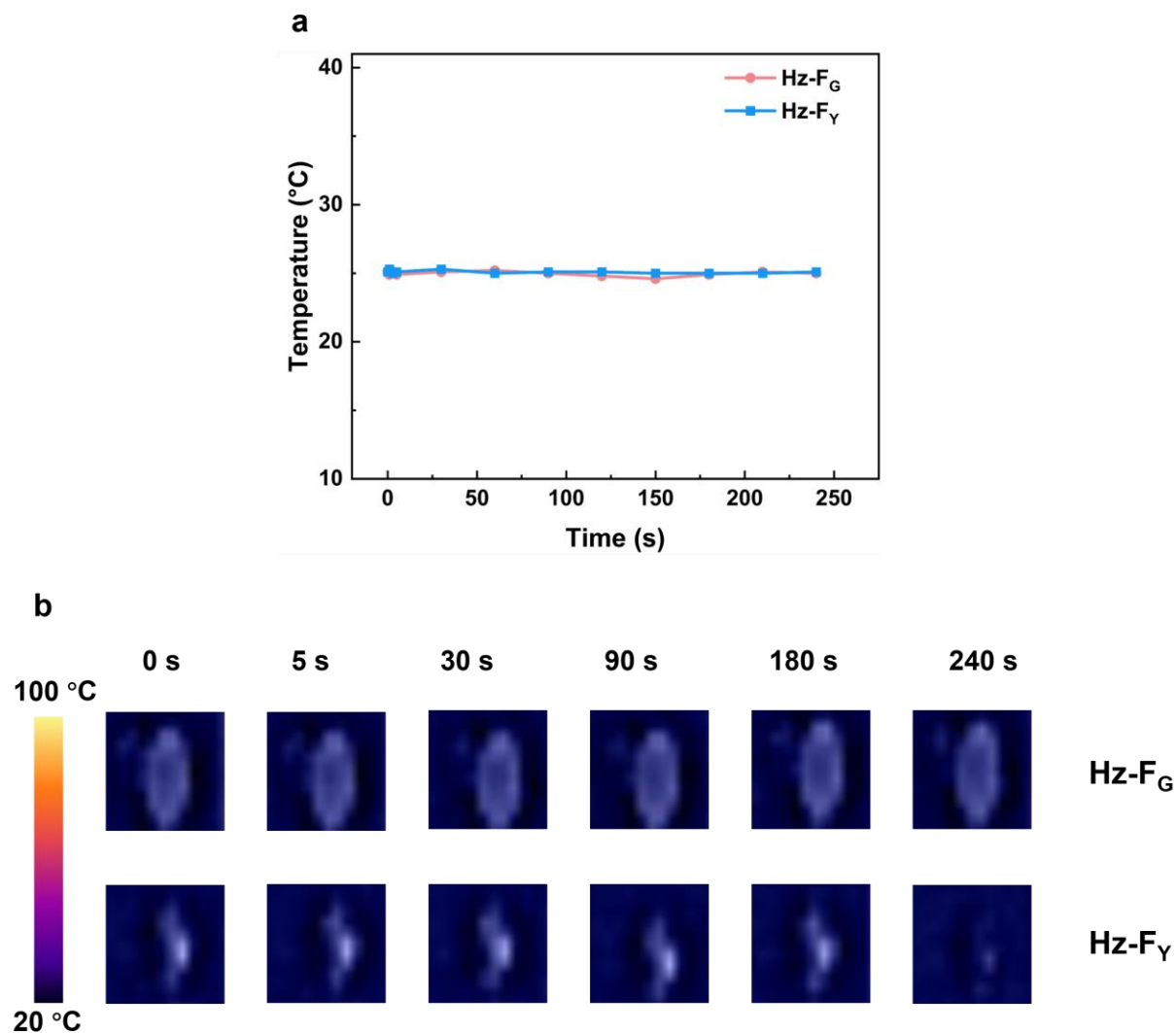


Figure S4. (a) Temperature variations of **Hz-F_G** and **Hz-F_Y** crystals under the irradiation of a 380 nm laser (1 mW). (b) Infrared thermal images recorded with a thermal imaging camera with different exposure time.

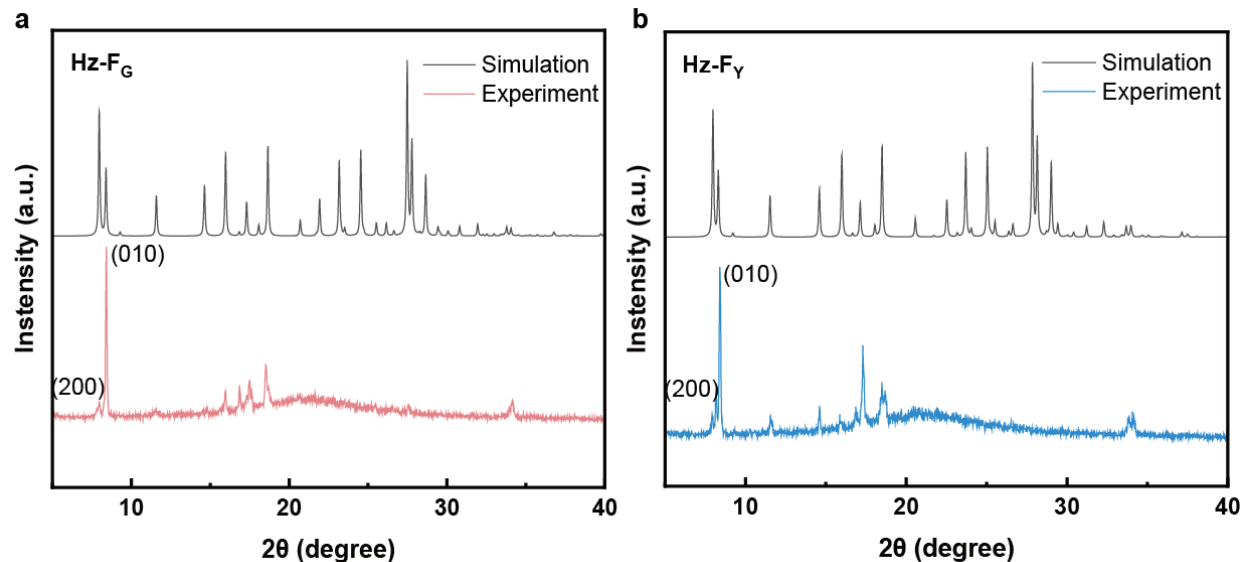


Figure S5. The PXRD patterns of (a) **Hz-F_G** and (b) **Hz-F_Y**.

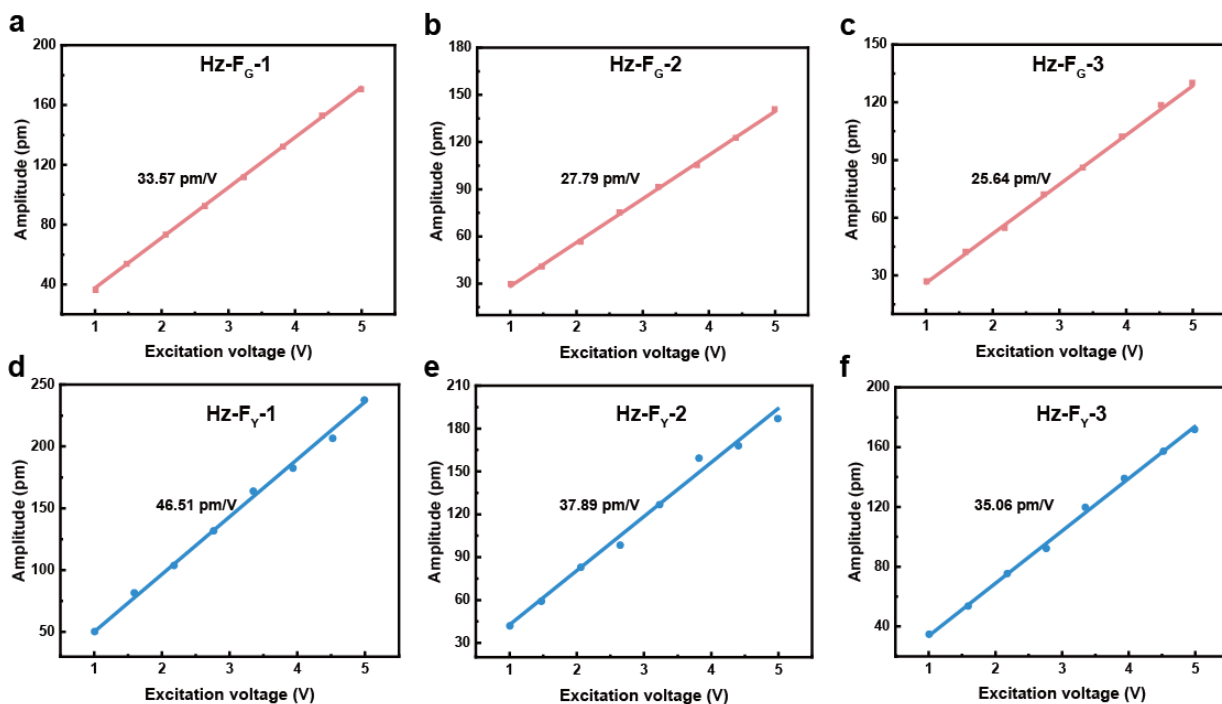


Figure S6. Quantitative piezoresponse measurements acquired on **Hz-F_G** and **Hz-F_Y** single crystals. Plots of piezoresponse amplitude versus AC drive voltage recorded from three distinct testing sites on (a-c) **Hz-F_G** and (d-f) **Hz-F_Y**. The slope of each curve corresponds to the local effective piezoelectric coefficient.

IV. Calculations

Table S3. Calculated piezoelectric charge tensor components e_{ij} (in units of C/m²), strain tensor components d_{ik} (pC/N), and elastic stiffness matrix c_{ij} (in units of GPa) of **H_z-F_G** and elastic compliance matrix components S_{ij} (GPa⁻¹).

H_z-F_G Charge Tensor components (C/m²)

$$\begin{pmatrix} 0.001 & -0.001 & 0 & -0.001 & -0.005 & -0.205 \\ 0 & 0 & 0 & 0 & -0.273 & -0.003 \\ -0.017 & 0.022 & 0.007 & 0 & 0 & 0 \end{pmatrix}$$

H_z-F_G Strain Tensor components (pC/N)

$$\begin{pmatrix} 0.047 & 0 & 0.579 & -0.063 & -2.13 & -41.9 \\ -0.052 & 0.15 & -0.147 & -0.167 & -49.754 & -0.693 \\ -5.975 & 7.39 & -6.525 & -0.023 & -0.009 & -0.028 \end{pmatrix}$$

H_z-F_G Elastic stiffness matrix (GPa)

$$\begin{pmatrix} 12.196 & 7.119 & -0.438 & 0 & 0 & 0 \\ 7.119 & 19.25 & 11.917 & 0 & 0 & 0 \\ -0.438 & 11.92 & 12.85 & 0 & 0 & 0 \\ 0 & 0 & 0 & 9.878 & 0 & 0 \\ 0 & 0 & 0 & 0 & 2.31 & 0 \\ 0 & 0 & 0 & 0 & 0 & 4.924 \end{pmatrix}$$

H_z-F_G Elastic compliance matrix components (GPa⁻¹)

$$\begin{pmatrix} 0.190 & -0.174 & 0.168 & 0 & 0 & 0 \\ -0.174 & 0.281 & -0.267 & 0 & 0 & 0 \\ 0.168 & -0.267 & 0.331 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0.101 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0.44 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0.203 \end{pmatrix}$$

Table S4. Calculated piezoelectric charge tensor components e_{ij} (in units of C/m²), strain tensor components d_{ik} (pC/N), and elastic stiffness matrix c_{ij} (in units of GPa) of **H₂-F₂** and elastic compliance matrix components S_{ij} (GPa⁻¹).

H₂-F₂ Charge Tensor components (C/m²)

$$\begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0.199 \\ 0 & 0 & 0 & 0 & 0.117 & 0 \\ 0.011 & -0.023 & 0 & 0 & 0 & 0 \end{pmatrix}$$

H₂-F₂ Strain Tensor components (pC/N)

$$\begin{pmatrix} 0.003 & -0.004 & 0 & 0 & 0 & 51.31 \\ 0 & 0.015 & 0 & 0 & 58.4 & 0 \\ 9.69 & -12.19 & 9.57 & 0 & 0 & 0 \end{pmatrix}$$

H₂-F₂ Elastic stiffness matrix (GPa)

$$\begin{pmatrix} 16.74 & 13.837 & 1.854 & 0 & 0 & 0 \\ 13.838 & 23.31 & 13.286 & 0 & 0 & 0 \\ 1.854 & 13.286 & 15.036 & 0 & 0 & 0 \\ 0 & 0 & 0 & 8.897 & 0 & 0 \\ 0 & 0 & 0 & 0 & 2.009 & 0 \\ 0 & 0 & 0 & 0 & 0 & 3.888 \end{pmatrix}$$

H₂-F₂ Elastic compliance matrix components (GPa⁻¹)

$$\begin{pmatrix} 0.27 & -0.28 & 0.22 & 0 & 0 & 0 \\ -0.30 & 0.39 & -0.31 & 0 & 0 & 0 \\ 0.22 & -0.31 & 0.31 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0.11 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0.50 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0.26 \end{pmatrix}$$

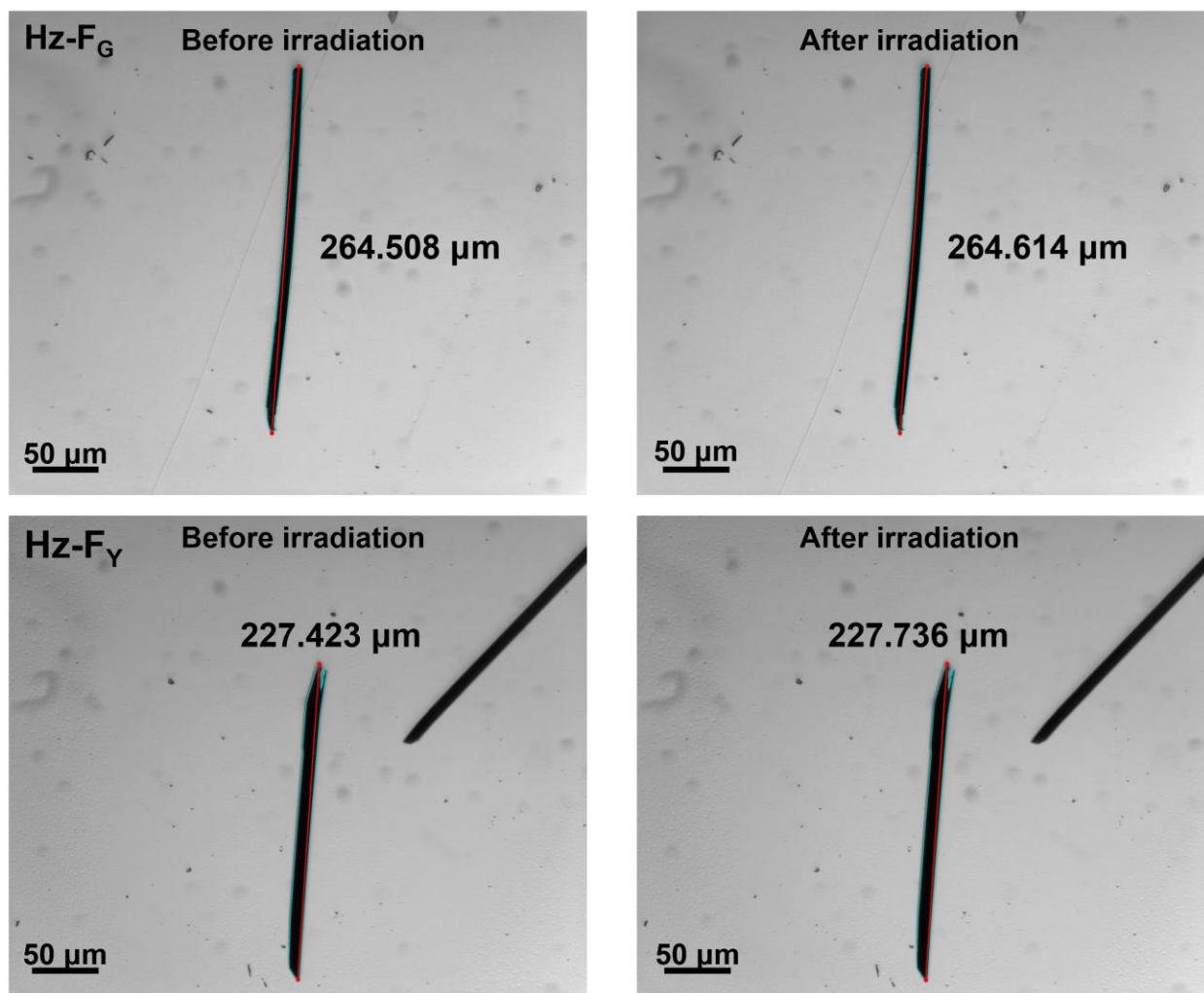


Figure S7. Representative optical microscopy images illustrating the photoinduced axial elongation of **Hz-F_G** and **Hz-F_Y** crystals upon laser irradiation. The left column displays the crystals before irradiation, whereas the right column shows them after irradiation. The crystal boundaries (green contours) and endpoints of the maximum Feret diameter (red points) were extracted via sub-pixel edge localization to determine the elongation of the crystals before and after irradiation.

Table S5. The lengths of **Hz-F_G** and **Hz-F_Y** crystals under 405 nm laser single-point irradiation and in dark.

	Initial crystal length L_0 [μm]	Crystal length after laser irradiation L_t [μm]	Elongation length $L_t - L_0$ [μm]	Average elongation length [μm]
Hz-F_G (1)	264.508	264.614	0.106	0.116 ± 0.018
	264.508	264.612	0.104	
	264.508	264.618	0.110	
	264.508	264.651	0.143	
Hz-F_G (2)	260.414	260.532	0.118	0.114 ± 0.006
	260.414	260.524	0.110	
	260.414	260.522	0.108	
	260.414	260.535	0.121	
	Initial crystal length L_0 [μm]	Crystal length after laser irradiation L_t [μm]	Elongation length $L_t - L_0$ [μm]	Average elongation length [μm]
Hz-F_Y (1)	230.410	230.635	0.225	0.227 ± 0.025
	230.410	230.605	0.195	
	230.410	230.666	0.256	
	230.410	230.644	0.234	
Hz-F_Y (2)	227.423	227.736	0.313	0.267 ± 0.040
	227.423	227.638	0.215	
	227.423	227.691	0.268	
	227.423	227.697	0.274	

V. Raman spectra

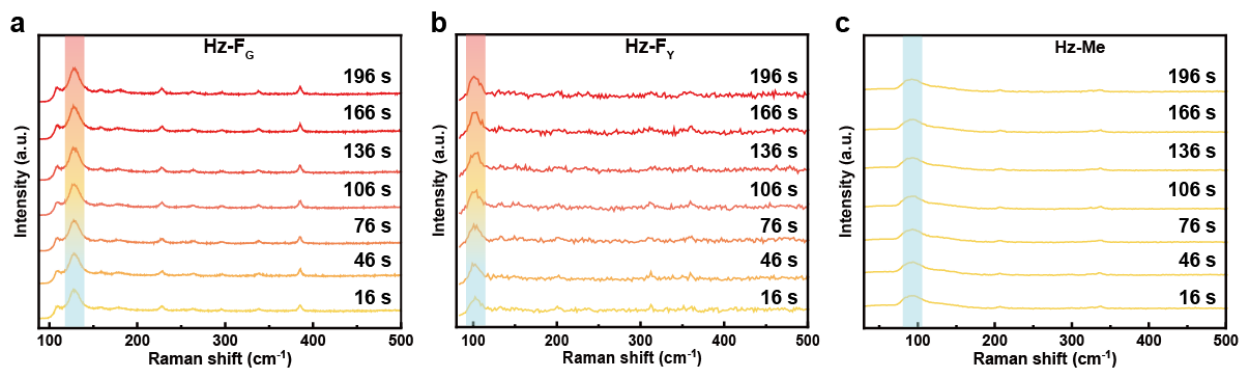


Figure S8. Lattice vibration peaks of (a) **Hz-F_G**, (b) **Hz-F_Y**, (c) **Hz-Me** single crystals from Raman spectroscopy.

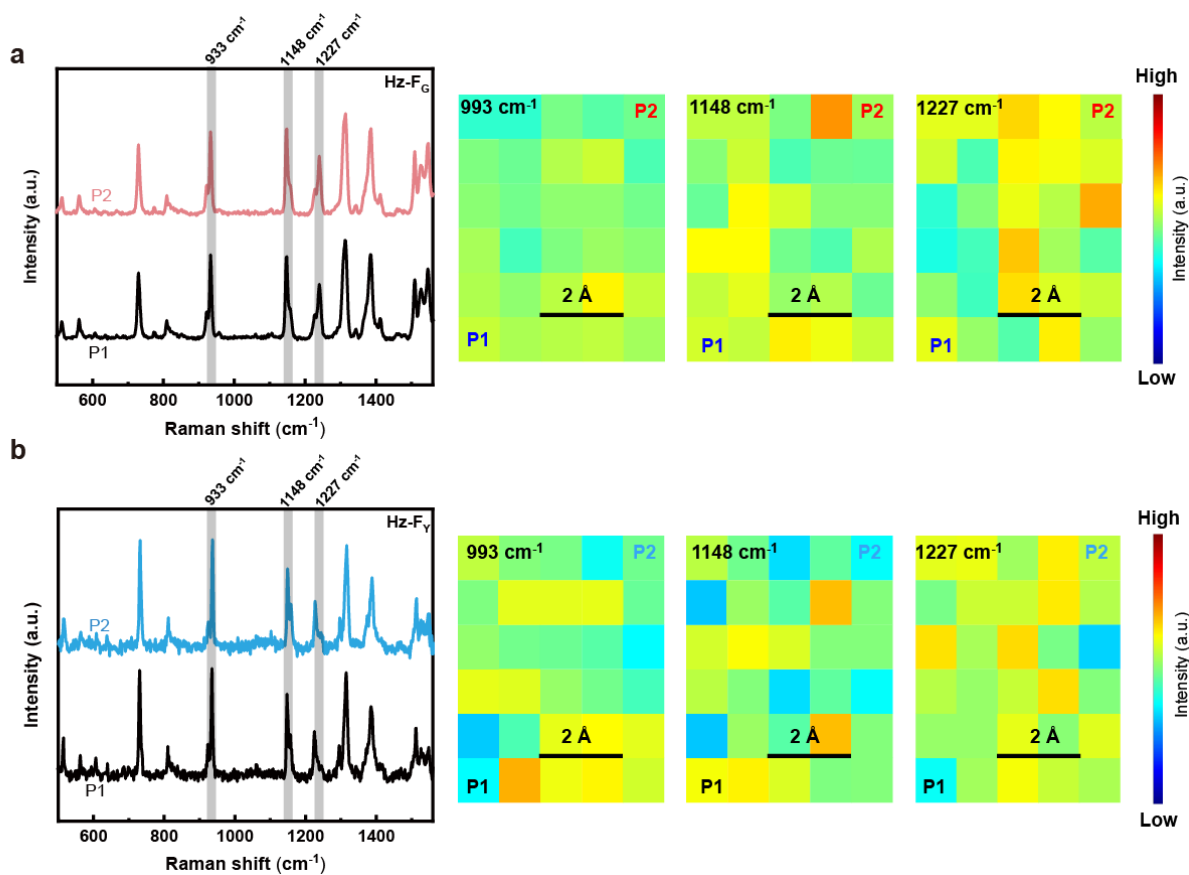


Figure S9. The Raman spectral peaks corresponding to the stretching and bending vibrations of chemical bonds in the aromatic regions of (a) **Hz-F_G** and (b) **Hz-F_Y** single crystals. On the right is the signal intensity mapping to the respective wave numbers.

VI. Elastic properties of crystals

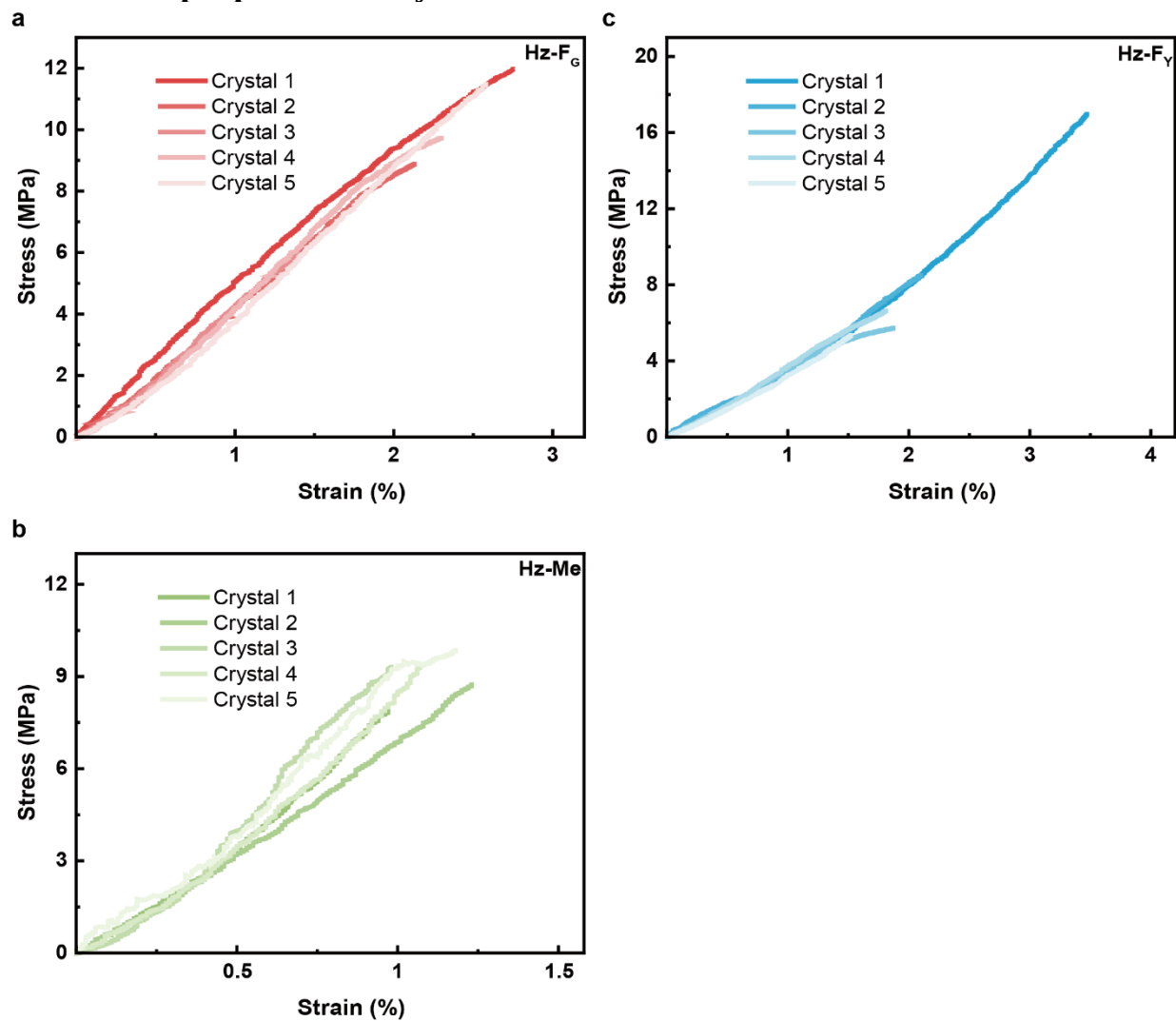


Figure S10. The stress-strain curves of the (a) **Hz-F_G**, (b) **Hz-F_Y** and (c) **Hz-Me** crystals.

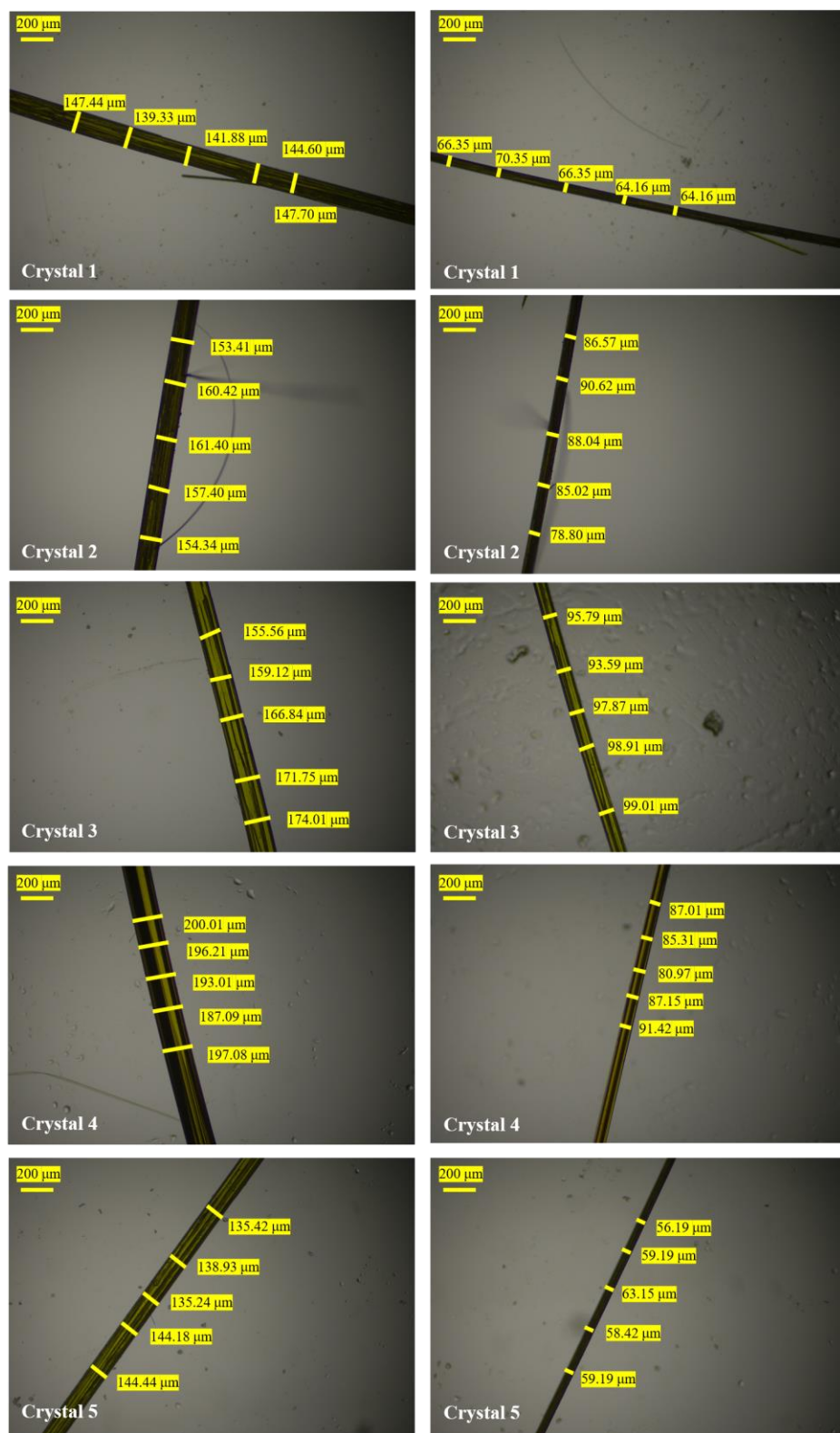


Figure S11. For each Hz-FG crystal subjected to tensile testing, the height and width were measured using microscopy. The cross-sectional area was determined from the average height and width values obtained across five representative segments per crystal.

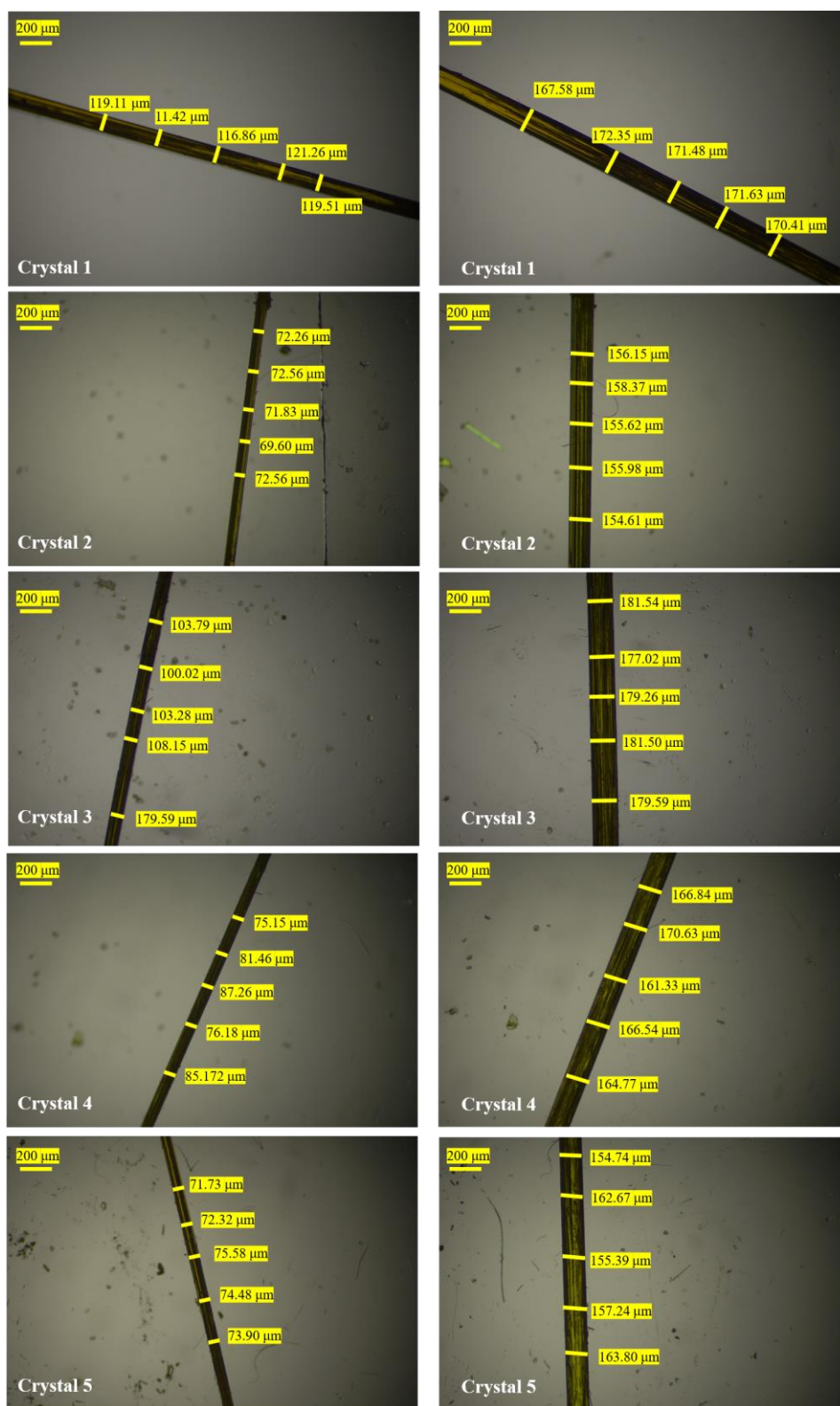


Figure S12. For each Hz-Fy crystal subjected to tensile testing, the height and width were measured using microscopy. The cross-sectional area was determined from the average height and width values obtained across five representative segments per crystal.

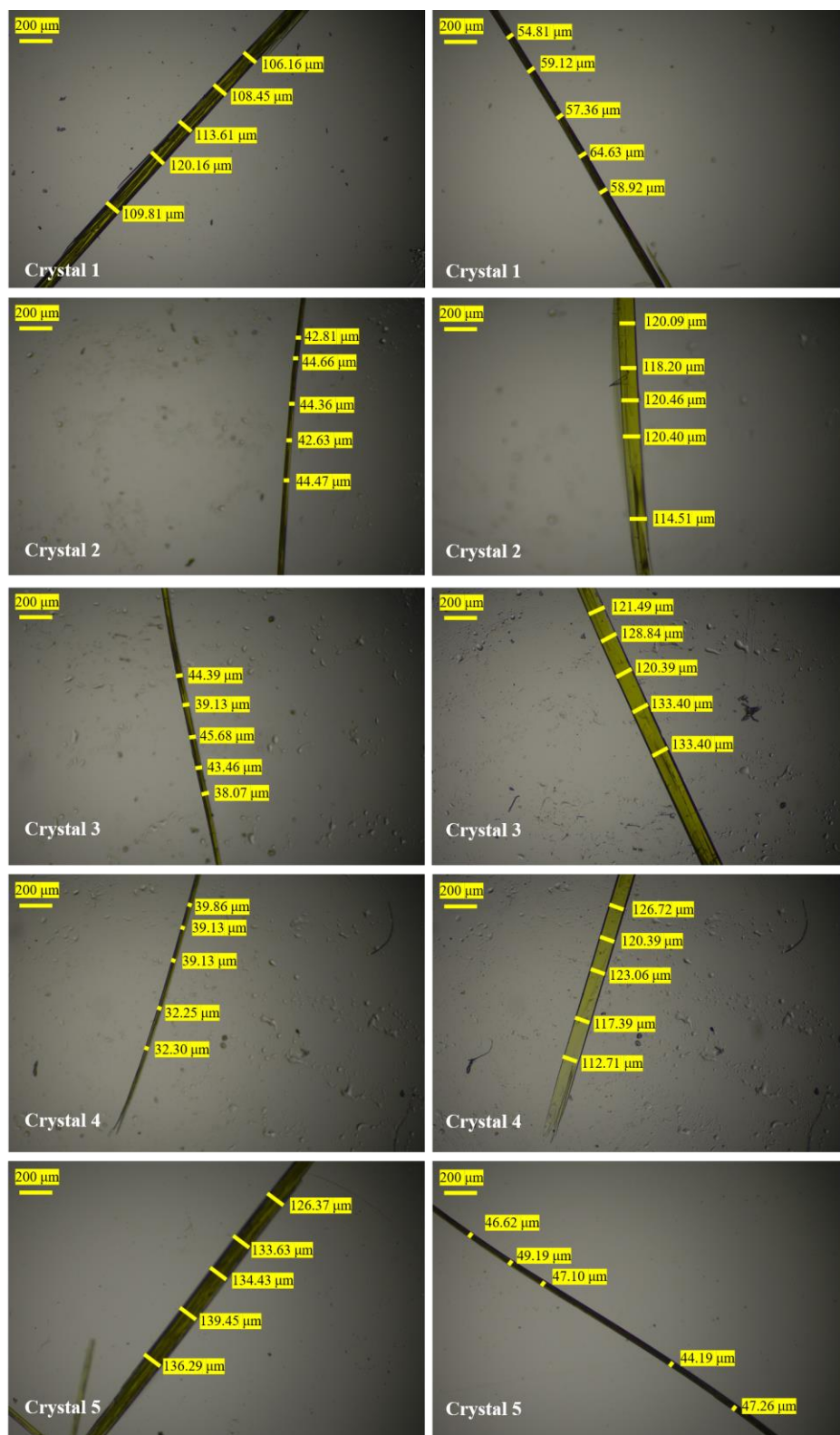


Figure S13. For each **Hz-Me** crystal subjected to tensile testing, the height and width were measured using microscopy. The cross-sectional area was determined from the average height and width values obtained across five representative segments per crystal.

Table S6. Statistical summary of the elastic moduli along the long-axis direction for **H_z-F_G**, **H_z-F_Y**, and **H_z-Me** molecular crystals.

Crystal	E (H_z-F_G) MPa	E (H_z-F_Y) MPa	E (H_z-Me) MPa
1	486	373	660
2	438	344	680
3	432	345	674
4	479	375	645
5	457	310	630
Average	458	349	658

Table S7. Calculated polycrystalline elastic parameters of **H_z-F_G** and **H_z-F_Y** crystals via the Voigt-Reuss-Hill approximation. The universal elastic anisotropy index (A^U) is employed to quantify elastic anisotropy, defined as:

$$A^U = 5 \frac{G_V}{G_R} + \frac{B_V}{B_R} - 6$$

B , Bulk modulus; G , Shear modulus; E , Young's modulus; ν , Poisson's ratio; A^U , Universal elastic anisotropy index. Subscripts V, R and H denote Voigt bound, Reuss bound and Hill average value, respectively. Moduli are given in units of GPa.

Parameter		H_z-F_G	H_z-F_Y
Bulk Modulus B (GPa)	B_V	9.06	12.56
	B_R	3.91	4.47
	B_H	6.48	8.52
Young's Modulus E (GPa)	E_V	12.10	12.53
	E_R	5.78	4.92
	E_H	9.37	8.73
Shear Modulus G (GPa)	G_V	5.14	4.70
	G_R	2.30	1.87
	G_H	3.72	3.28
Poisson's ratio ν	ν_V	0.26	0.33
	ν_R	0.25	0.32
	ν_H	0.26	0.33
A^U		7.33	9.38

VII. Crystal data and theoretical calculations

Table S8. Crystal data and structure refinement for **Hz-F_G**, **Hz-F_Y** and **Hz-Me**.

Parameters	Hz-F _G	Hz-F _Y	Hz-Me
Empirical formula	C ₂₄ H ₁₂ F ₃ N ₇	C ₂₄ H ₁₂ F ₃ N ₇	C _{13.50} H _{10.50} N _{3.50}
Formula weight	455.41	455.41	221.75
Temperature/K	150.0	150.0	100.0
Crystal system	orthorhombic	orthorhombic	orthorhombic
Space group	Pmn2 ₁	Pmn2 ₁	Pnma
a/Å	22.1509 (18)	22.17 (2)	23.1304 (8)
b/Å	10.6369 (9)	10.522 (10)	23.0938 (8)
c/Å	4.0087 (3)	4.119 (4)	3.971784 (11)
α/°	90	90	90
β/°	90	90	90
γ/°	90	90	90
Volume/Å ³	944.52 (13)	960.8 (16)	2121.63 (12)
Z	2	2	8
ρ _{calc} (g/cm ³)	1.601	1.574	1.388
μ/mm ⁻¹	0.122	0.120	0.689
F (000)	464.0	464.0	928.0
Radiation	MoKα (λ = 0.71073)	MoKα (λ = 0.71073)	CuKα (λ = 1.54184)
2θ range for data collection/°	4.248 to 54.962	2.143 to 27.537	3.82 to 76.035
Index ranges	-28 ≤ h ≤ 28, -13 ≤ k ≤ 13, -4 ≤ l ≤ 5	-28 ≤ h ≤ 20, -13 ≤ k ≤ 13, -4 ≤ l ≤ 5	-28 ≤ h ≤ 28, -21 ≤ k ≤ 21, -4 ≤ l ≤ 4
Reflections collected	7539	4379	14411
Independent reflections	2159 [R _{int} = 0.0706, R _{sigma} = 0.0660]	2117 [R _{int} = 0.0733, R _{sigma} = 0.0969]	1967 [R _{int} = 0.0772, R _{sigma} = 0.0533]
Data/restraints/parameters	2159/1/163	2117/1/163	1967/0/166
Goodness-of-fit on F ²	1.042	1.022	1.056
Final R indexes [I ≥ 2σ (I)]	R ₁ = 0.0523, wR ₂ = 0.1212	R ₁ = 0.0626, wR ₂ = 0.1376	R ₁ = 0.0461, wR ₂ = 0.1187
Final R indexes [all data]	R ₁ = 0.0900, wR ₂ = 0.1431	R ₁ = 0.1362, wR ₂ = 0.1772	R ₁ = 0.0577, wR ₂ = 0.1265
Largest diff. peak/hole / e Å ⁻³	0.34/-0.27	0.333/-0.253	0.217/-0.208

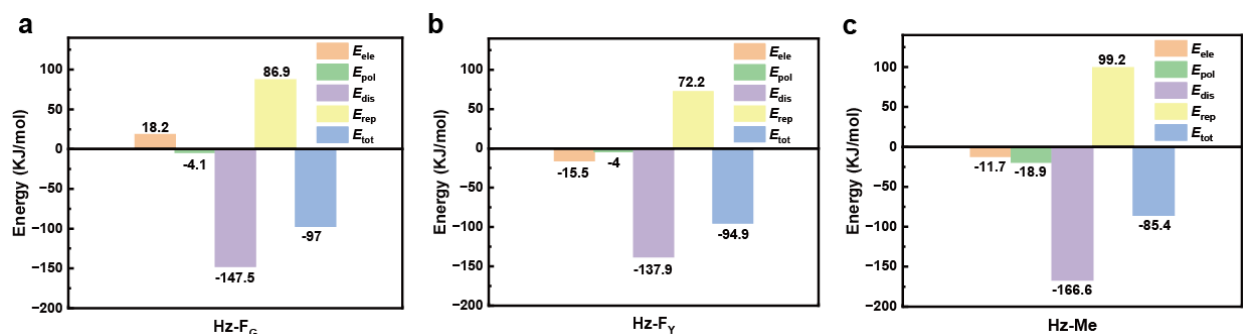


Figure S14. Energy decomposition analysis of intermolecular interactions in **Hz-R** crystals. Intermolecular interaction energies are partitioned into electrostatic energy (E_{ele}), polarization energy (E_{pol}), dispersion energy (E_{dis}), exchange-repulsion energy (E_{rep}), and total interaction energy (E_{tot}). Panels correspond to (a) **Hz-F_G**, (b) **Hz-F_Y**, (c) **Hz-Me**. All energies are reported in units of kJ mol^{-1} .

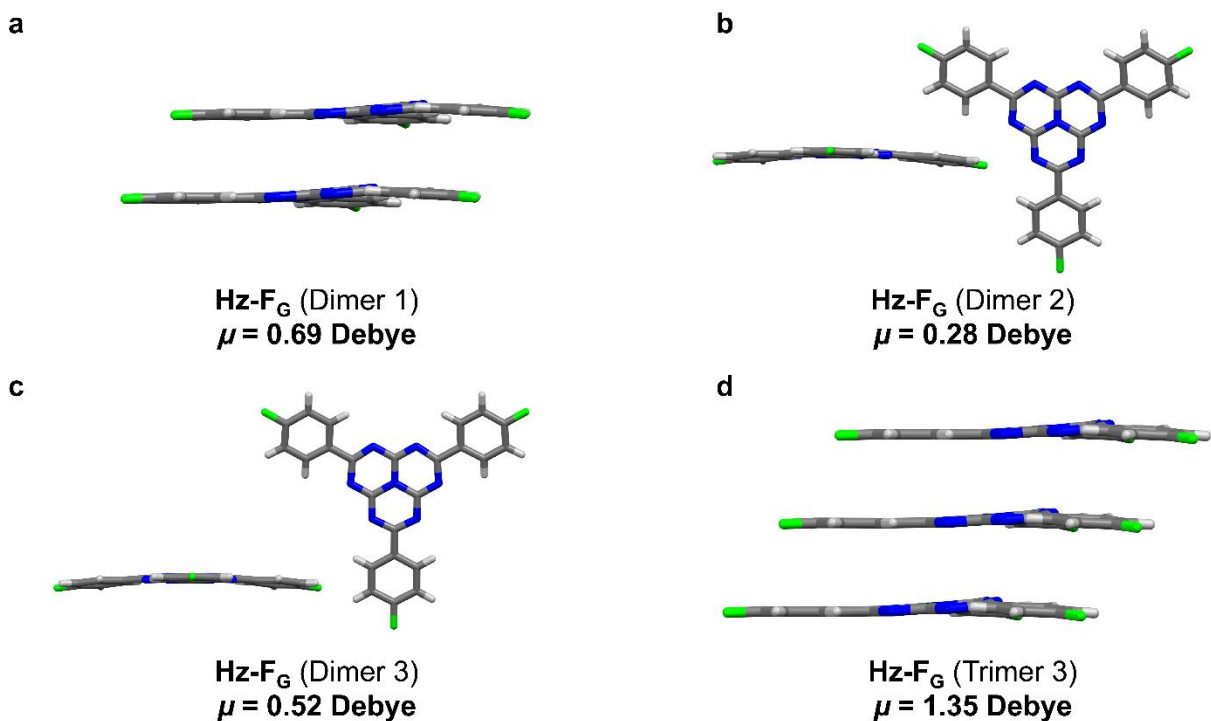


Figure S15. (a-d) Dipole moments of dimers and trimers in **Hz-F_G** crystals.

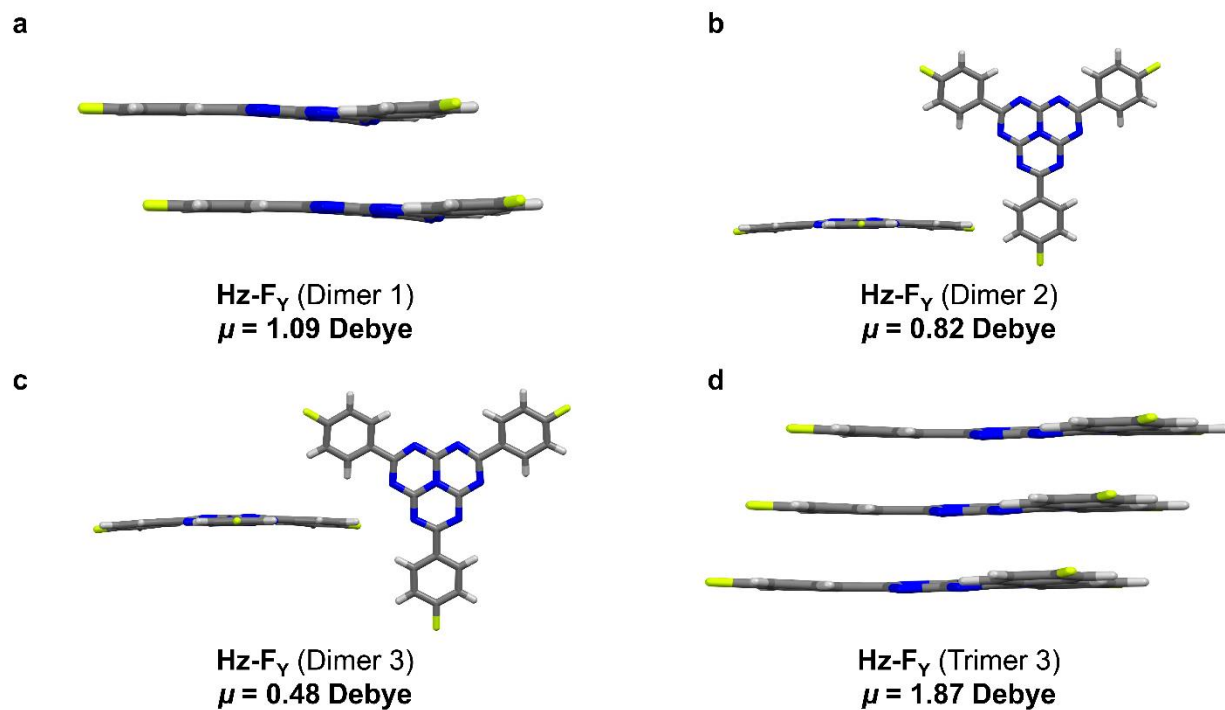


Figure S16. (a-d) Dipole moments of dimers and trimers in **Hz-F_Y** crystals.

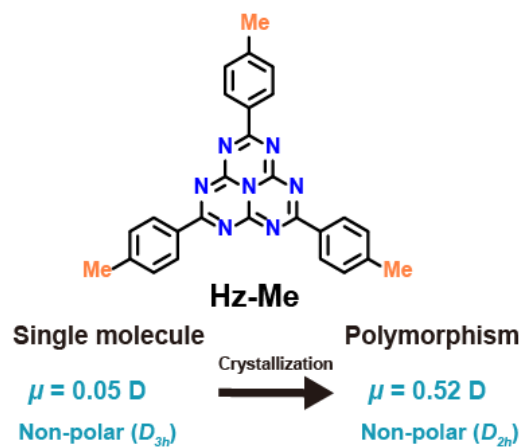


Figure S17. Molecular structure of **Hz-Me** and symmetry evolution from single molecule to polymorphism.

VIII. Mechanism of crystal motion

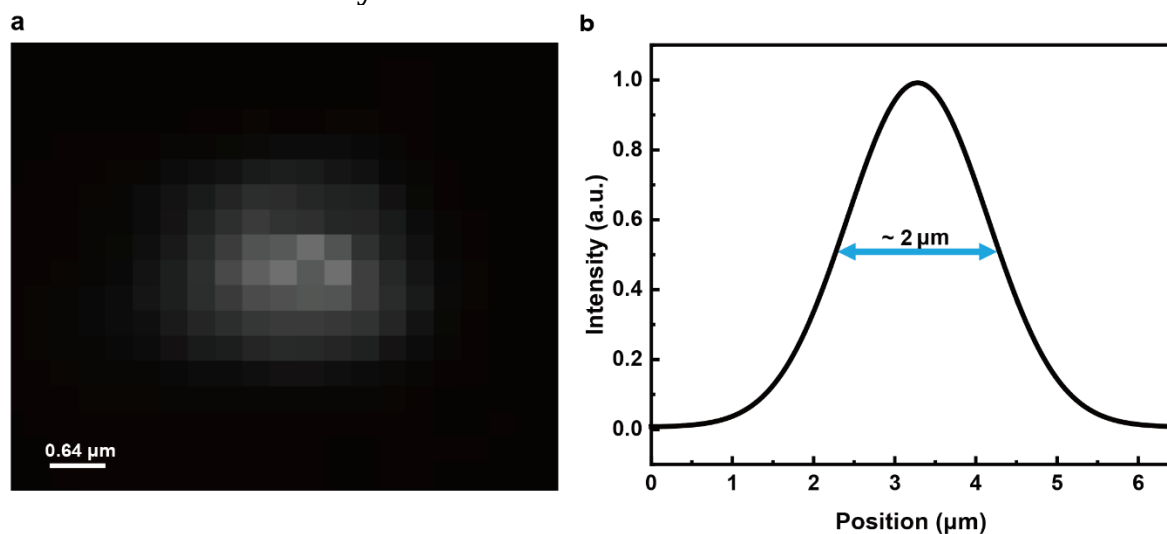


Figure S18. (a) The laser spot photograph taken by a CCD camera under a $20\times$ objective lens. (b) The spot diameter is defined using the full width at half maximum (FWHM), yielding a diameter of approximately $2\ \mu\text{m}$.

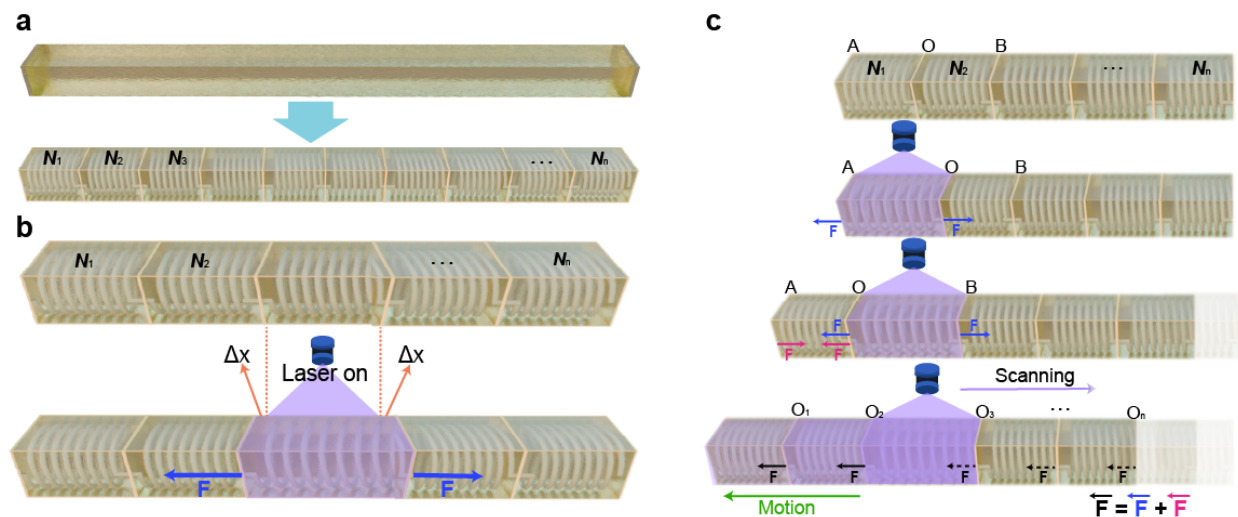


Figure S19. Mechanism of light-induced antiparallel motion based on coupling between a segmented spring model and photo-triggered local elastic deformation. (a) The crystal is divided into N identical ideal independent linear spring segments along the c -axis. (b) A single segment undergoes symmetric axial elastic stretching under localized laser illumination. (c) Continuous laser scanning drives macroscopic displacement of the crystal.

IX. ^1H NMR spectra

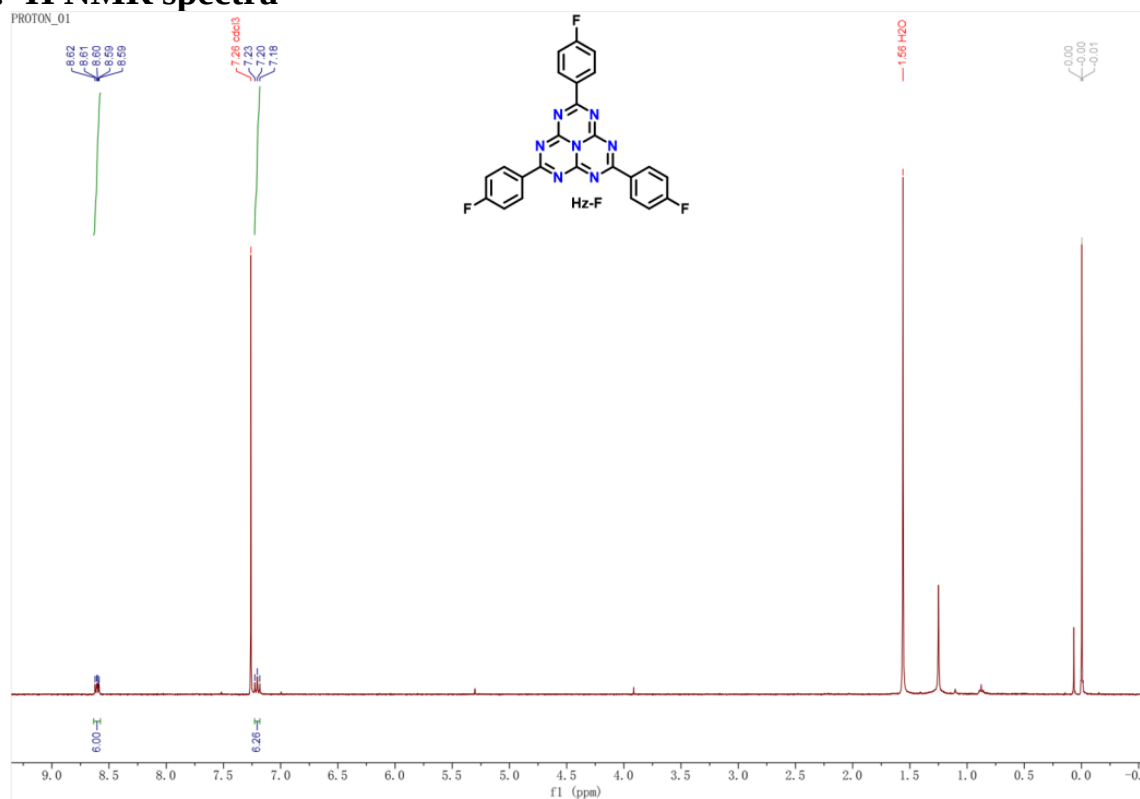


Figure S20. ^1H NMR spectrum of **Hz-F**.

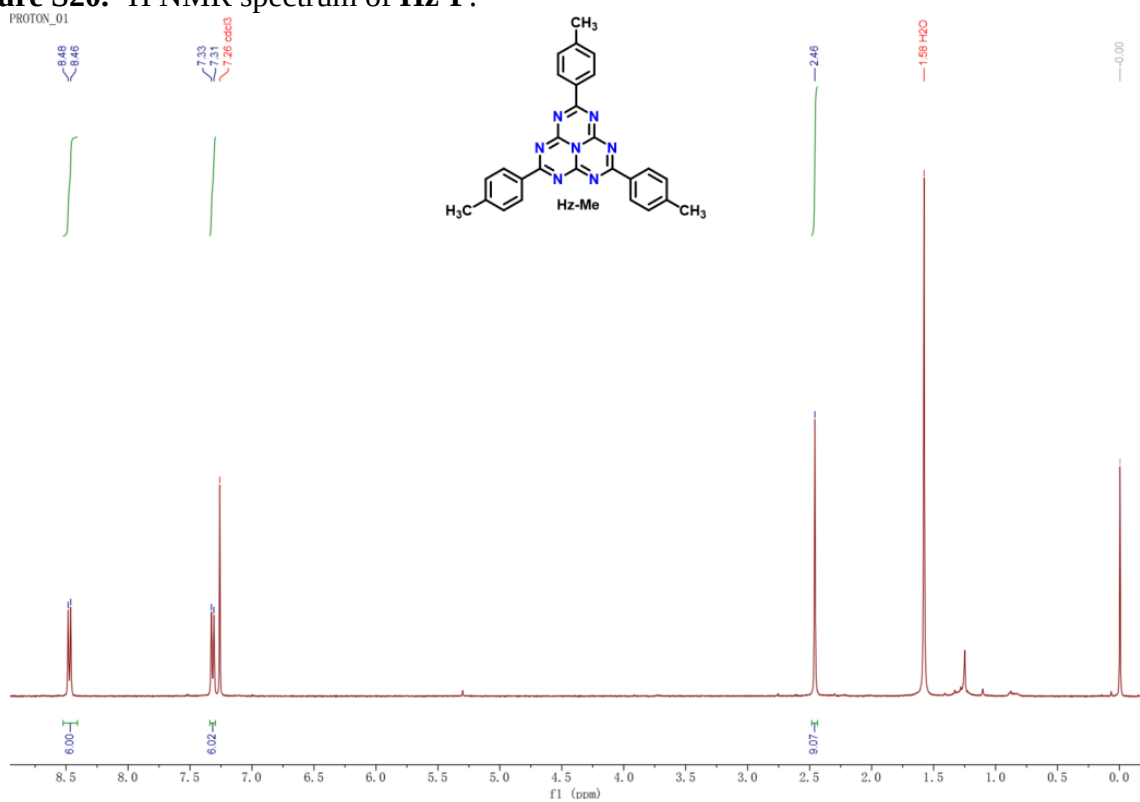


Figure S21. ^1H NMR spectrum of **Hz-Me**.

X. References

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