Supporting Information

Molecular Mechanisms of Superelasticity and Ferroelasticity in Organic Semiconductor Crystals

Hong Sun[1], Sang Kyu Park[2], Ying Diao[2], Eric P. Kvam[3*], Kejie Zhao[1*]

[1]School of Mechanical Engineering, Purdue University, West Lafayette, Indiana 47907, United States.

[2]Department of Chemical and Biomolecular Engineering, University of Illinois Urbana—Champaign, 600 S. Mathews Avenue, Urbana, Illinois 61801, United States.

[3]School of Materials Engineering, Purdue University, West Lafayette, Indiana 47907, United States.

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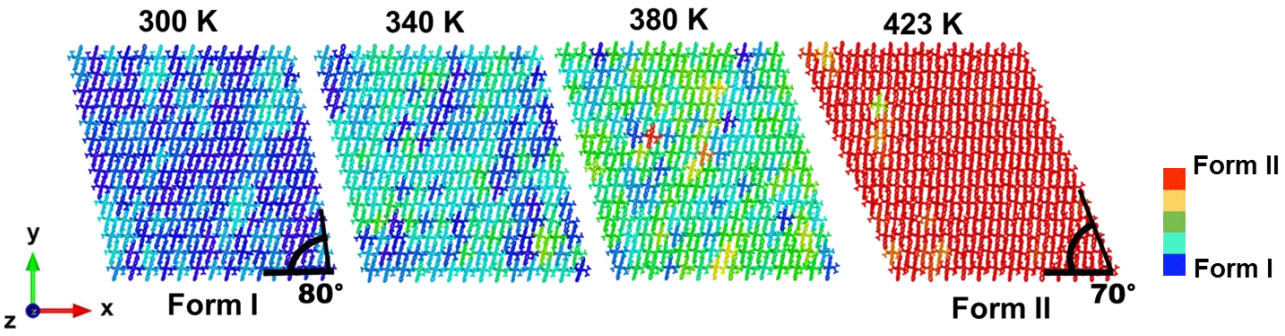

Figure S1. Molecular trajectories of Form I-to-II transition upon heating.

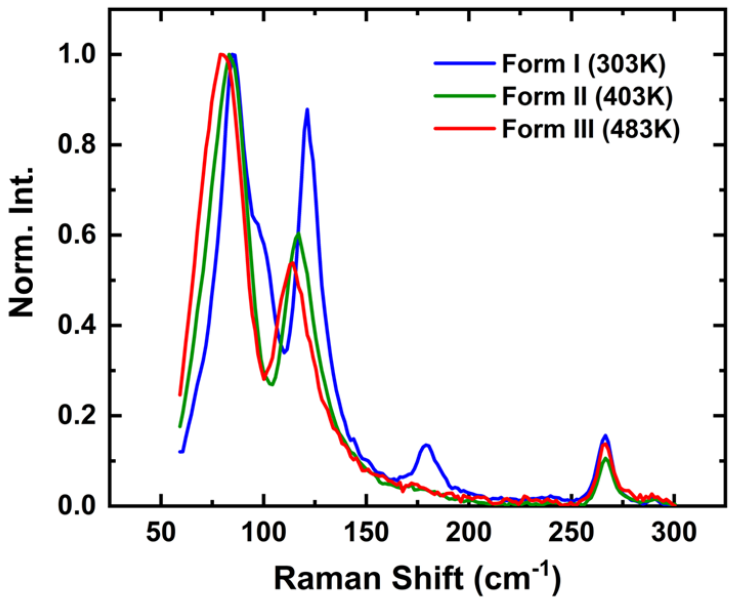

Figure S2. Raman spectrum for different polymorphs under their equilibrium temperatures.

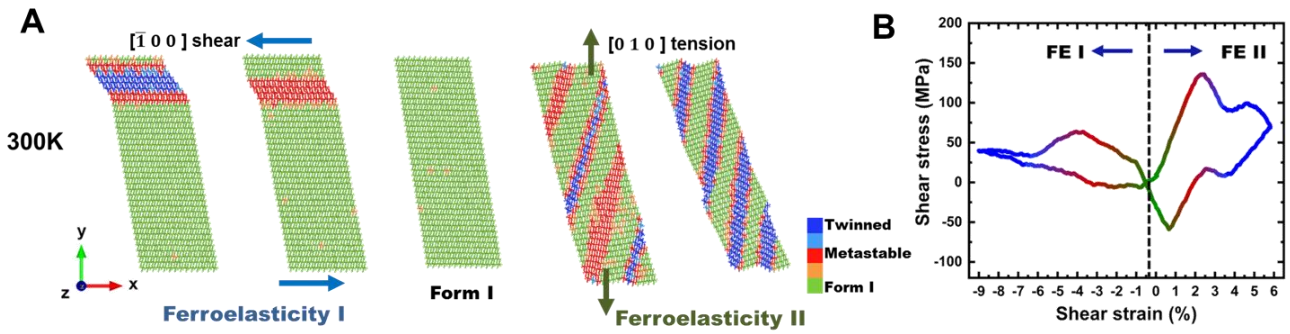

Figure S3. (A) The molecular trajectories of FE-I and FE-II of Form I by [$\bar{1}$ 0 0] shear and [0 1 0] tension at 300K, respectively. Their corresponding stress-strain responses are shown in (B). The molecules in (A) are color-coded by the order parameter representing the different phases as marked in the color bars.

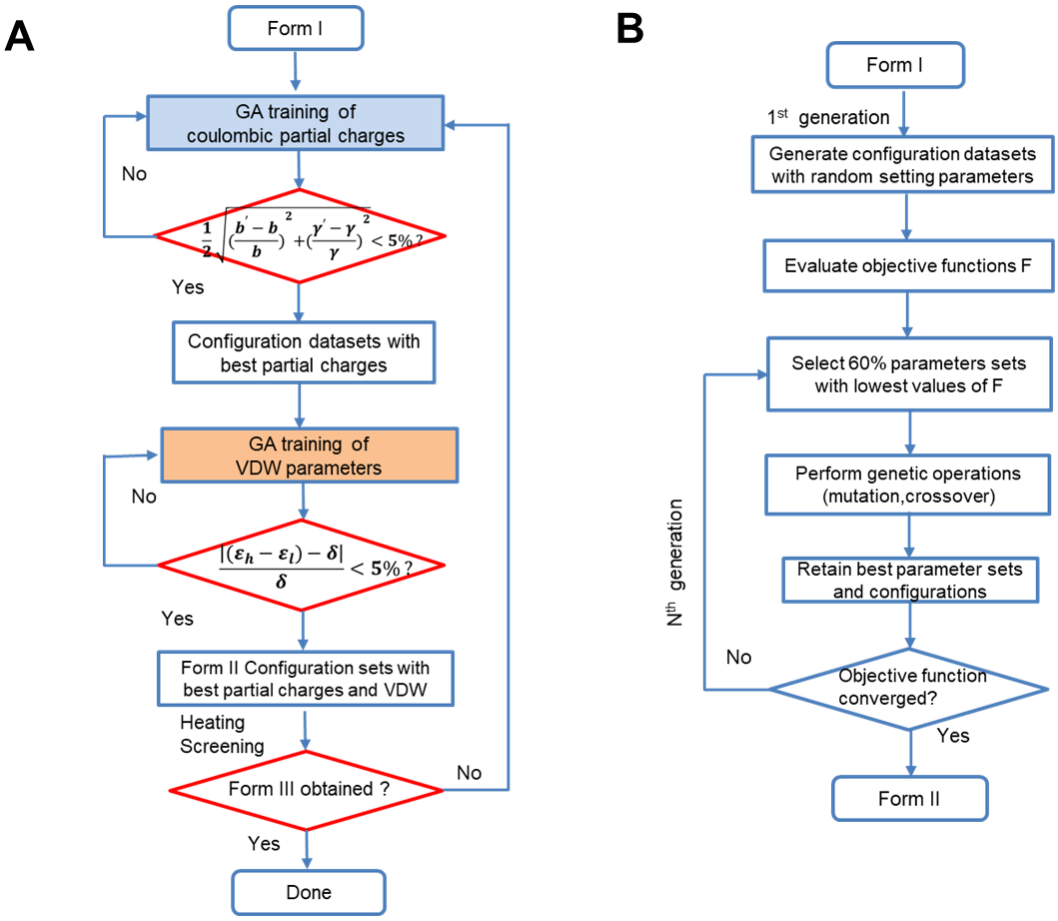

Figure S4. (A) Flowchart of parametric training for coulombic and van der Waals parameters. (B) The workflow of the Genetic-Algorithm training module.

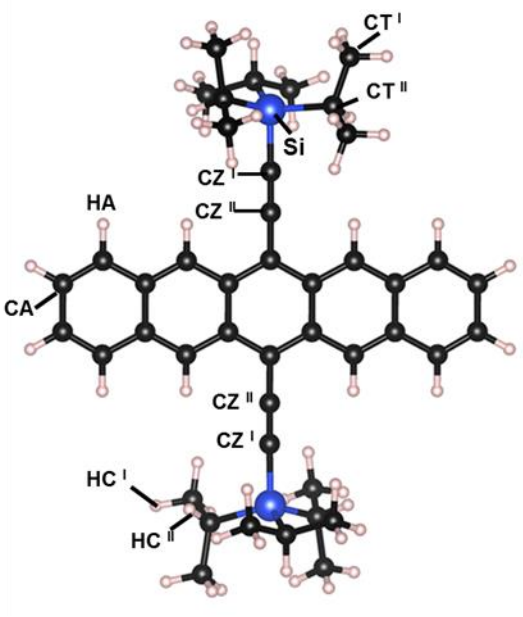

Figure S5. The atomic symbol of OPLS-AA force field assigned to each atom in TIPS-P molecule. **CA** and **HA** are for carbon atom and hydrogen atom of pentacene backbone, respectively. **CT^{I}** and **HC^{I}** are for carbon atom and hydrogen atom of the methyl group in the TIPS unit, respectively. **CT^{II}** is for carbon atom connecting Si atom and two methyl groups in the TIPS unit. **HC^{II}** is for hydrogen atom bounded to **CT^{II}** atom. **CZ^{I}** and **CZ^{II}** are for the *sp* carbon atoms.

Table S1. The simulated lattice parameters (red) agree well with the experimental data (blue). Their percentage differences are shown in the parentheses.

	a	b	c	α	β	γ
Form I	7.76	7.74	16.95	77.8	88.5	82.2
	7.64	7.87	17.13	74.5	86.6	80.2
	(-1.5%)	(+1.7%)	(+1.0%)	(-4.3%)	(-2.2%)	(-2.4%)
⇩						
Form II	7.60	8.58	17.23	78.2	86.7	72.1
	7.76	8.54	17.69	71.0	82.0	70.6
	(+2%)	(-0.4%)	(+2%)	(-9%)	(-5%)	(-2%)
⇩						
Form III	7.60	8.99	17.27	82.2	89.9	65.2
	8.00	8.98	17.88	81.0	88.3	65.5
	(+5%)	(-0.1%)	(+3.5%)	(-1.4%)	(-1.8%)	(+0.5%)

Table S2. Coulombic partial charges for TIPS-pentacene molecules.

Atomic symbol	Atomic charge (e)	Atomic type in OPLS-AA
CT^{I}	0.0448	Alkane CH3-
CT^{II}	0.1611	Alkyl Silane R2CH-Si
CZ^{I}	0.1659	Alkyne RCCR
CZ^{II}	-0.0194	
CA	0.0754	Aromatic C
HA	-0.1685	Aromatic H-C
HC^{I}	-0.055	Alkane H-C
HC^{II}	0.0595	
Si	0.0938	Alkyl Silane R4Si

Table S3. van der Waals parameters for TIPS-pentacene molecules.

Atomic symbol	Depth of potential well (Kcal/mol)	Lennard-Jones radii (Å)	Atomic type in OPLS-AA
CT	0.05957	3.5	Alkane CH3-
CZ	0.2005	3.3	Alkyne RCCR
CA	0.06383	3.55	Aromatic C
HA	0.03066	2.42	Aromatic H-C
HC	0.03580	2.5	Alkane H-C
Si	0.09025	4.0	Alkyl Silane R4Si