Supplementary Materials for

Internal strain tunes electronic correlations on the nanoscale

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DOI: 10.1126/sciadv.aau9123

This PDF file includes:

Section S1. Sample preparation
Section S2. Optical far-field experiments
Section S3. Landau free energy calculations
Fig. S1. AFM profile of the sample surface.
Fig. S2. Far-field reflectivity on different spots of the cracked sample.
Fig. S3. Temperature-dependent far-field experiments.
Fig. S4. Schematics of the geometry used to treat the elasticity problem.
Section S1. Sample preparation

Flat α-(BEDT-TTF)$_2$I$_3$ single crystals with homogeneous surface were first selected on the basis of their visual appearance. The material properties were checked by polarized reflectance measurements, in order to verify the proper phase, orientation, etc. Atomic force microscopy (AFM) was employed to ensure that the as-grown sample surface is of adequate quality for near-field experiments. Specimens fulfilling all these requirements were then mounted on commercial sapphire substrates and the contacts were glued using conductive carbon paste.

We deposited a gold layer of 30 - 40 nm thickness with an edge of less than 1 µm width, which is shown in the AFM images in fig. S1. These maps were acquired in situ, simultaneously with optical near-field measurements at low temperature around the phase transition ($T_{CO} = 136$ K). In panel A one can nicely see the flat surface of the homogeneous single crystal which is

![AFM profile of the sample surface.](image)

**Fig. S1. AFM profile of the sample surface.** (A) The homogeneous single crystal shows a flat surface with roughness of a few nm only. The line profile indicates the evaporated gold layer (‘Au’) with an edge of less than 1 µm. (B) The cracked sample consists of many loosely connected segments with appreciable difference in height $h$. The surface is more strongly distorted than in (A), which certainly correlates with internal strain.

investigated in Fig. 2 of the main paper. On the other hand, the cracked sample shown in Fig. 3 consists of many single-crystalline segments which are loosely connected via small edges due to the large internal strain, as shown in fig. S1 B.
Section S2. Optical far-field experiments

Subsequent to cryogenic near-field experiments, we performed polarized reflection measurements on the cracked sample. The use of an infrared microscope Bruker Hyperion allows us to selectively access different areas on the crystal surface with a minimum resolution of $\approx 30 \times 30 \, \mu \text{m}^2$. The reflectivity measured on four different spots, shown in fig. S2, reveals the typical anisotropy and spectral characteristics of $\alpha$-(BEDT-TTF)$_2$I$_3$ (17). At frequencies above the plasma edge ($\omega > 4000 \, \text{cm}^{-1}$) the crystal becomes transparent due to weak absorption and small thickness. The resulting Fabry-Perot oscillations have similar periodicity for all spots,

![Graphs showing reflectivity vs. frequency for different spots on the cracked sample.](image)

**Fig. S2. Far-field reflectivity on different spots of the cracked sample.** The polarization-dependent infrared spectra reveal the characteristic response of the two in-plane crystal axes (17). The Fabry-Perot fringes, most pronounced at high frequencies, have similar periodicity on different positions on the surface indicating that the crystal has a uniform thickness of 15 $\mu$m. The differences in absolute value stem from different alignment of the particular segments with respect to the incoming light.

implying a uniform thickness of $d \approx 15 \, \mu$m on the entire sample. Depending on the alignment of a segment with respect to the incoming light, the absolute value of the reflected intensity is different and the Fabry-Perot fringes are pronounced more or less strongly. Figure S3 A-C shows photographs of the sample surface and the respective spots investigated by temperature
sweeps; note the scale bars. Spot 1 corresponds to the area quantitatively evaluated in the main manuscript, as indicated in panel D. In order to evaluate the temperature-dependent changes of the optical conductivity with high temperature resolution, the temperature was ramped with 0.01 K/min through the charge-order transition and the reflectivity was measured in 0.01 K intervals. As illustrated in fig. S3 E, the reflectivity gradually decreases with cooling. The false-color plot (panel F) infers that the most pronounced changes happen around 134 – 135 K. The overall intensity is quantified by integrating the spectral weight $SW(T)$ between $\omega_1$ and $\omega_2$ at each temperature, which is plotted for spots 1 – 4 in fig. S3 H. Likewise, the largest rate of change is illustrated by the derivative $dSW(T)/dT$ in panel G. We have chosen the interval 700 - 1000 cm$^{-1}$ since the spectral response at low frequencies is most susceptible to discern between metallic/insulating behavior due to the opening of the charge order gap (17).

The temperature evolution of the far-field intensity on spot 1 develops in a way similar to the overall near-field signal. In other words, the far-field measurement sums up the spatially distributed response of all metallic and insulating regions, resulting in a gradual change rather than an abrupt jump as one would expect for a first-order transition. The largest portion of pixels changes phase when the absolute intensity is subject to its most pronounced changes. For spot 1 and 3 the behavior is very similar; so it is safe to assume similar phase coexistence. On the other hand, the transition is significantly broader on spot 2, with largest rate of change at slightly lower temperature. In the context of our main results, this region is, most likely, subject to more pronounced internal strain causing a larger temperature range of coexistence. The situation is even more delicate on spot 4 where the transition is interrupted. Due to its location in the corner of a segment, close to a point where several cracks join, this indicates the coexistence of two distinct regions. The first one undergoes the metal-insulator transition around 134 K while charge order is suppressed at 130 K and below in the second one, reminiscent of much stronger strain. Due to limited resolution of our far-field setup, however, we cannot rule out a small crack in the monitored region, separating it into two thermally isolated segments with a significant temperature offset.
**Fig. S3. Temperature-dependent far-field experiments.** (A) Room temperature image of the multiply-cracked α-(BEDT-TTF)$_2$I$_3$ single crystal (‘sample 3’). (B) The cracks appear at low temperatures. (C) Colored squares indicate the spots where far-field reflectivity measurements were performed. (D) The red and blue areas in the near-field map (cf. Figure 3) correspond to spots 1 and 2, respectively. (E) The low-frequency optical reflectivity is substantially reduced upon cooling through the charge-order transition (measured in 0.01 K steps; here shown for spot 1), which is illustrated in the false-color plot (F). To analyze the phase transition more quantitatively, the reflectivity was integrated between $\omega_1 = 700$ cm$^{-1}$ and $\omega_2 = 1000$ cm$^{-1}$ to cover the frequency range of most pronounced changes. (H) The resulting spectral weight $SW(T)$ was normalized to the high- and low-temperature plateau values, i.e. deep in the metallic and charge-ordered phases, respectively. After similar normalization, the temperature dependence of the average near-field intensity agrees perfectly with the far-field data of spot 1 (cf. Figure 3). (G) The derivative with respect to temperature highlights the largest rate of change. Apparently, the transition does not happen at the same temperature on the different spots. While it is very similar and sharp for spots 1 and 3, it is much broader on spot 2. For region 4 it even seems to be interrupted. To rule out systematic errors, we performed a few more measurements on spot 1 after looking at the different spots, giving reproducible results.
To conclude, bulk probes with a diffraction-limited spatial resolution, such as far-field optical spectroscopy utilizing an infrared microscope, are sensitive to phase coexistence and phase inhomogeneities as well; yet, it would be difficult to come to the same conclusions without the precision and high-quality data of the near-field technique.

Section S3. Landau free energy calculations

Elastic energy density of a phase-change crystal.

Here we elaborate on the Landau theory proposed in the main text and, moreover, the pursuant simulations of the strain-coupled charge-ordering phase transition in α-(BEDT-TTF)₂I₃

\[ F = \int_V dV \left( f_{CO}(\vec{r}) + f_\varepsilon(\vec{r}) + f_D(\vec{r}) \right) \]  

(Eq. S1)

with

\[ f_T \equiv \frac{1}{2} k |\nabla \psi|^2 + \alpha (T - T_{CO}) \psi^2 - \beta \psi^3 + \gamma \psi^4, \]

\[ f_\varepsilon \equiv \frac{1}{2} \left[ \varepsilon^* \psi(\vec{r}) - \varepsilon(\vec{r}) \right] : K : \varepsilon^* \psi(\vec{r}) \]

and

\[ f_D \equiv D(\vec{r}) \psi(\vec{r}) \]

The coupling of the order parameter \( \psi(\vec{r}) \) to the local strain field \( \varepsilon(\vec{r}) \) can be understood as consequence of a structural transformation of the unit cell upon the transition from the metallic to the insulating charge-ordered (CO) phase, giving rise to a so-called “misfit strain” (also called “spontaneous strain”) \( \varepsilon^* \) (47). In this case, the total elastic energy of the system is given by

\[ F_\varepsilon = \frac{1}{2} \int_V dV \left( \varepsilon(\vec{r}) - \varepsilon^* \psi(\vec{r}) \right) : K : \left( \varepsilon(\vec{r}) - \varepsilon^* \psi(\vec{r}) \right) \]

where

\[ \varepsilon^*_{ij} \equiv \frac{\Delta \alpha_i}{\alpha_i} \delta_{ij} \]  

(Eq. S2)

The integration volume \( V \) extends from the surface of the crystal (\( z = 0 \)) throughout its entire depth (\( z < 0 \)). Here \( \Delta \alpha_i \) signifies the change in lattice constant \( \alpha_i \) (\( i = a, b, c \)) upon entering the CO phase, in which \( \psi(\vec{r}) > 0 \); \( K \) denotes the rank-four stiffness tensor, according to which we define the stresses \( \varepsilon \equiv K : \varepsilon \) and \( \varepsilon^* \equiv K : \varepsilon^* \); and \( \delta_{ij} \) denotes the Kronecker delta. We emphasize here that \( \varepsilon(\vec{r}) \) quantifies the strain relative to atomic coordinates “natural” to the \( L \) phase, in which elastic energy vanishes when \( \varepsilon = 0 \). For simplicity in the present application, we consider α-(BEDT-TTF)₂I₃ as an elastically isotropic solid for which elements of the stiffness tensor are given by

\[ K_{ijkl} = \Lambda \delta_{ij} \delta_{kl} + 2 \mu \frac{1}{2} (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) \]

where \( \Lambda \) and \( \mu \) are the material’s Lamé and shear moduli, respectively (49). Relations between these moduli and the Young’s modulus \( E \) (taken to be \( \approx 10^{11} \) dynes/cm\(^3\) based on values for similar organic crystals (35)) and
Poisson’s ratio $\nu$ (assumed $\approx 0.3$) are available from standard texts of elasticity theory \((49)\).

We briefly highlight the geometry considered for application of the present Landau theory, as depicted in fig. S4. We consider a cross-section of a $\alpha$-(BEDT-TTF)$_2$I$_3$ crystal slab as glued to the sapphire substrate, in particular one of those segments subjected to spontaneous fracture while cooled to low temperature (see main text). Here the $x$-$z$ cross-sectional plane aligns with the $a$-$c$ plane of the crystal, while the dimensions of the crystal segment are $L$ in the $x$-direction and $D$ in the $z$-(thickness)direction. For comparison with data presented in the main text, we consider cracked boundaries of the crystal parallel to the $b$-direction, imparting (at least locally) a translational symmetry to the system in this direction. Therefore, for simplicity, we consider our model system to have translational symmetry in this direction.

Now, we note that 1) $\varepsilon(\vec{r}) \equiv \nabla \vec{u}(\vec{r})$, with $\vec{u}$ the vector field of atomic displacements, and that 2) mechanical equilibrium requires vanishing $\mathcal{S}(\vec{r}) - \mathcal{S}^* \psi(\vec{r})$ at the exposed (free) boundaries of the crystal at $x=0, L$ where the crystal has fractured to relieve thermoelastic stress, as well as the crystal top surface at $z=D$ (see fig. S4). We denote these free boundaries by $\Omega_{\text{free}}$. The bottom boundary $z=0$ of the crystal volume $V$ we take to be clamped (“glued”) in place upon the substrate and we denote this boundary region $\Omega_{\text{clamped}}$. The implicit assumption for this boundary condition is that sapphire has a much higher Young’s modulus (is much “stiffer”) than $\alpha$-(BEDT-TTF)$_2$I$_3$; this convenient simplification allows to consider only elastic fields within the crystal itself. These considerations allow simplification of Eq. S2 by the divergence theorem, under which we obtain a surface integral contribution ($\int d\mathcal{S} \cdot ...$)

$$F_\varepsilon = \frac{1}{2} \left[ \int_{\Omega_{\text{clamped}}} d\mathcal{S} \cdot \left( \mathcal{S}(\vec{r}) - \mathcal{S}^* \psi(\vec{r}) \right) \cdot \vec{u}(\vec{r}) \right]$$

$$- \int_V dV \varepsilon : \left( \mathcal{E}(\vec{r}) - \mathcal{E}^* \psi(\vec{r}) \right)$$

$$- \int_V dV \left( \varepsilon^* \psi(\vec{r}) - \mathcal{E}^* \psi(\vec{r}) \right) : \sigma^* \psi(\vec{r}) \right] \quad \text{(Eq. S3)}$$

Meanwhile, we can discard the surface integral under the assumption that the sample is clamped at the substrate with infinite rigidity, under which condition here $\vec{u} = \vec{0}$. We next note that a term $\varepsilon^* : \mathcal{S}^* \psi(\vec{r})^2$ simply modifies the Landau free energy density contribution scaling with $\psi^2$, which in the case of our Landau theory amounts to no more than an effective shift of the critical temperature $T_{CO}$. Therefore, we give this term no further consideration. Subsequent discussion will show that the first volume integral of Eq. S3 will vanish. The remaining contribution to $F_\varepsilon$ reflects linear coupling between the order parameter and strain, as hypothesized and presented in Eq. 2 of the main text.
\[ F_e = \frac{1}{2} \int_V dV \, \mathbf{E}(\vec{r}) : \mathbf{S}^* \psi(\vec{r}) \]
\[ = \frac{1}{2} \left[ \int_V dV \, \mathbf{u}(\vec{r}) \cdot \nabla \left( \mathbf{S}^* \psi(\vec{r}) \right) - \int_{\Omega_{\text{free}}} dS \dot{z} \cdot \left( \mathbf{S}^* \mathbf{u}(\vec{r}) \right) \psi(\vec{r}) \right] \quad \text{(Eq. S4)} \]

Here we have applied integration by parts and discarded contributions to the surface integral at clamped boundaries where \( \mathbf{u} = 0 \), excepting the free boundaries.

We now consider functional minimization \( F_e \) with respect to \( \mathbf{u}(\vec{r}) \). This is equivalent to the condition of mechanical equilibrium, which requires

\[ \nabla \cdot \left( \mathbf{S}(\vec{r}) - \mathbf{S}^* \psi(\vec{r}) \right) = -\bar{f}(\vec{r}) \quad \text{(Eq. S5)} \]

with \( \bar{f} \) the local body force density. In absence of explicit defects or internal forces \( \bar{f} = 0 \) and thus

\[ \nabla \cdot \mathbf{S}(\vec{r}) = \nabla \cdot \left( \mathbf{S}^* \psi(\vec{r}) \right) \quad \text{(Eq. S6)} \]

In this sense, when considering the strain field \( \mathbf{E}(\vec{r}) \), the quantity \( -\nabla \left( \mathbf{S}^* \psi(\vec{r}) \right) \) represents an internal pseudo-body force. Consequently, the first volume integral (second term) of Eq. S3 indeed vanishes. Moreover, mechanical equilibrium at crystal boundaries requires

\[ \dot{z} \cdot \left( \mathbf{S}(\vec{r}) - \mathbf{S}^* \psi(\vec{r}) \right) = -\bar{T}(\vec{r}) \quad \text{(Eq. S7)} \]

with \( \bar{T} \) the surface traction. Since the unclamped crystal boundaries are elastically unconstrained (traction-free), here \( \bar{T} = 0 \), and thus

\[ \dot{z} \cdot \mathbf{S}(\vec{r}) = \dot{z} \cdot \mathbf{S}^* \psi(\vec{r}) \equiv \bar{S}_z \psi(\vec{r}) \quad \text{(Eq. S8)} \]

Therefore, the quantity \( -\bar{S}_z \psi(\vec{r}) \) represents a pseudo-surface traction.

Under these criteria \( \mathbf{u}(\vec{r}) \) may be determined up to a constant as the solution to Navier’s equation \(^3\) under the pseudo-body force and pseudo-surface traction profiles established by \( \nabla \psi(\vec{r}) \) and \( \psi(\vec{r}) \), respectively
\[ \tilde{u}(\vec{r}) = \tilde{u}_{\text{therm}}(\vec{r}) + \tilde{u}_\psi(\vec{r}), \quad \text{and} \]
\[ \tilde{u}_\psi(\vec{r}) = \int_V d\vec{r}' G_f(\vec{r} - \vec{r}') \left( - \nabla \left( \hat{S}^* \psi(\vec{r}') \right) \right) + \int_{\vec{r}'=0} dS' \hat{G}_T(\vec{r} - \vec{r}') \left( - \hat{S}_z^* \psi(\vec{r}') \right) \quad \text{(Eq. S9)} \]

First, \( \tilde{u}_{\text{therm}} \) is a \( \psi \)-independent solution to the homogeneous Navier’s equation, namely \( \nabla \cdot \left( K : \nabla \tilde{u}_{\text{therm}} \right) = 0 \), under crystal boundary conditions imposed by thermoelastic strain, i.e. differential thermal contraction of the \( \alpha \)-(BEDT-TTF)$_2$I$_3$ compared with the sapphire substrate at a temperature \( T \approx 150 \text{K} \). In particular, this contribution to \( \tilde{u} \) satisfies the extrinsic boundary condition \( E_{xx} \approx -0.0075 \) at \( z = H \) and associates with the “background” thermoelastic strain field shown in Fig. 4F of the main text. As mentioned, boundary conditions on the other crystal surfaces still satisfy the traction-free condition \( \hat{n} \cdot \left( K : \nabla \tilde{u}_0 \right) = 0 \), with \( \hat{n} \) the outwards-facing unit vector. According to these boundary conditions, we compute \( \tilde{u}_{\text{therm}} \) with the finite element method in the quasi-2D geometry of the \( xz \)-plane shown in Fig. S4.

Next, \( \tilde{u}_\psi(\vec{r}) \) associates with inhomogeneities in the order parameter \( \psi \) which energetically predispose the crystal to a state of inhomogeneous stress according to Eq. S9. Here \( G_f \) and \( G_T \) are Green’s dyadic tensors that solve Navier’s equation\(^3\) for the elastic semi-infinite half-space in the presence of a point-like (“delta Dirac”) body force and surface traction, respectively, each originating at some location \( \vec{r}' \) in the \( xz \)-plane. The point-inhomogeneous dyadic \( G_f \) satisfies the traction-free boundary condition \( \hat{t} \cdot \hat{n} G_f(\vec{r}) = \vec{t} = 0 \) at \( z = 0 \), whereas \( G_T \) is homogeneous in the crystal volume: \( \nabla \cdot \left[ K : \nabla \left( \hat{\beta} \cdot G_T(\vec{r}) \right) \right] = \vec{f} = 0 \) in \( V \), for \( \hat{\beta} = \hat{x}, \hat{y}, \hat{z} \) corresponding to all possible orientations of surface traction. These dyadics are analytically available through standard solution methods\(^3\), but their qualitative long-range behavior is the most essential feature, as we later discuss. Moreover, \( \xi_0 \) is a constant of integration that can in principle range from 0 to \( \xi^* \), thus accommodating a net expansion or contraction of a totally unclamped crystal for the removal of elastic energy; however, our choice of clamped boundary condition at the substrate implies \( \xi^* = 0 \) here, and consequently only internal displacement fields \( \tilde{u}(\vec{r}) \) vanishing at the crystal’s clamped boundary are permitted. For completeness, we note that the finite extent of a parallelepiped crystal volume up to clamped \( \tilde{u} = 0 \) boundaries is expected to modify the “semi-infinite” dyadics through the addition of canceling “image charge” contributions from positions across the crystal boundaries. Although these exact “image dyadics” are used for calculations in the present work, their detailed description is not relevant for qualitative features of our results; these details will be described in a forthcoming publication.
Insertion of the Eq. S9 solution into Eq. S4 reveals that volumetric elastic energy density \( f_\varepsilon(\vec{r}) \) effectively consists of long-range strain-mediated interactions between Eshelby-type inclusions \((50)\) distributed according to the profile of \( \psi(\vec{r}) \)

\[
f_\varepsilon(\vec{r}) = -\frac{1}{2} \left[ \int_V dV' \nabla \left( \bar{S}^* \psi(\vec{r}') \right) \mathcal{G}_T(\vec{r} - \vec{r}') \nabla \left( \bar{S}^* \psi(\vec{r}') \right) \right] + 2 \int_{z'=0} dA' \nabla \left( \bar{S}^* \psi(\vec{r}') \right) \mathcal{G}_T(\vec{r} - \vec{r}') \bar{S}^*_z \psi(\vec{r}') + \mathcal{E}_{\text{therm}}(\vec{r}) : \bar{S}^* \psi(\vec{r}) \tag{Eq. S10}
\]

To obtain the doubled second term within square brackets, we have applied the fact that \( \mathcal{G}_T(\vec{r} - \vec{r}') = \mathcal{G}_T(\vec{r}' - \vec{r}) \). Here \( \mathcal{E}_{\text{therm}}(\vec{r}) \) is the “background” thermoelastic strain field (see Fig. 4F of the main text) associated with the pre-computed displacement field \( \vec{u}_{\text{therm}}(\vec{r}) \). For completeness, we also denote by \( \partial F_\varepsilon(\vec{r}) \) the elastic energy per unit surface area associated exclusively with the free surface of the crystal

\[
\partial F_\varepsilon(\vec{r}) = -\frac{1}{2} \int_{z'=0} dS \bar{S}^*_z \psi(\vec{r}) \mathcal{G}_T(\vec{r} - \vec{r}') \bar{S}^*_z \psi(\vec{r}') \tag{Eq. S11}
\]

Because \( \mathcal{G}_T(\vec{r} - \vec{r}') \) is long-range, this contribution has the qualitative effect of offsetting the aforementioend effective decrease in critical temperature \( T_{CO} \) proportionate to \( \mathcal{E}^*_z : \bar{S}^* \) in a fashion scaling with the area of crystal surface free to expand or contract. Therefore, we give \( \partial F_\varepsilon(\vec{r}) \) no further consideration for our present task, since it plays no direct role in the energetics that drive spatial heterogeneity through the charge-order phase transition. Finally, Eq. S10 now shows explicitly that the thermoelastic strain field behaves like a “linear-coupling” field that energetically “encourages” charge ordering whenever \( \mathcal{E}_{\text{therm}} : \bar{S}^* > 0 \); that is, wherever the local strain distorts the unit cell towards dimensions of the charge-ordered unit cell. This is the qualitative means by which the transition “nucleates” at the fractured (free) ends of the thermoelastically distorted crystal.
Fig. S4. Schematics of the geometry used to treat the elasticity problem. The bottom surface (red) of the crystal is considered clamped by adhesive to the (assumed) rigid sapphire substrate. The $a$-axis oriented ($x=0, L$) boundaries of the crystal have cracked away from the parent crystal due to thermoelastic strain induced by differential thermal expansion with the substrate. These cracked boundaries, the top surface, and interior volume of the crystal are all considered free to develop spontaneous strain that minimizes elastic energy during spatial coexistence of metallic and charge-ordered phases through the charge-ordering transition.

**Dimension**

To simplify the problem at hand from a three-dimensional to a quasi two-dimensional system, we consider the translational symmetry along the $b$-direction endowed by our model system. Under this simplification, $\psi$ becomes a function $\psi(x,z)$ of the cross-sectional coordinates $x$ and $z$, volume integrals reduce to surface integrals in the $xz$-plane multiplied by a $b$-oriented depth $t$ into the crystal slab, and $G_f(r-r')$ corresponds with the displacement field resulting from a point-columnar body force oriented within the $xz$-plane and extending a distance $t$.

**Phenomenology of the strain-induced striped texture.**

In the main part of the paper we only give a heuristic explanation for the formation of a striped texture of the order parameter within the regime of phase coexistence for a strain-coupled first-order transition. Here we provide a more detailed explanation anchored in the reformulation of elastic energies in terms of long-range strain-mediated interactions between “inclusions” of $\psi(\vec{r})$ (Eq. S10). The immediately relevant contributions $f_{stripe}$ to the Landau free energy (Eq. 1 of the
main text) are the following

\[
 f_{\text{stripe}}(\vec{r}) = \frac{1}{2} \left[ \kappa |\nabla \psi|^2 - \int_V dV' \nabla \cdot \left( S^* \psi(\vec{r}) \right) G_f (\vec{r} - \vec{r}') \nabla \left( S^* \psi(\vec{r}') \right) \right] \tag{Eq. S12}
\]

Eq. S12 should be interpreted as a competition between 1) the reduction of elastic energy through formation of abundant domain boundaries (\(|\nabla \psi| > 0\)), and 2) the energetic penalty of order parameter “stiffness” \(\kappa\). If we strictly consider \(z\)-independent and spatially periodic forms for \(\psi(\vec{r})\), e.g. at in-plane wave-vector \(\vec{k}\), then the free energy has the following form

\[
 F_{\text{stripe}} = \frac{1}{2} \int dA \left[ t \kappa |\nabla \psi|^2 - \int_V dA' \nabla \cdot \left( S^* \psi(\vec{r}) \right) \langle G_f \rangle_f (\vec{r} - \vec{r}') \nabla \left( S^* \psi(\vec{r}') \right) \right] \tag{Eq. S13}
\]

Here \(t\) is the thickness of the phase coexistent region along the \(b\)-axis of the crystal where \(\nabla \psi\) is nonzero, and \(\langle G_f \rangle_f \equiv \int_0^t dz \int_0^t dz' G_f\) represents an “effective” lateral interaction kernel. It suffices for the present discussion to consider the long-range behavior of \(\langle G_f \rangle_f\)

\[
 \langle G_f \rangle_f (\vec{r}) \sim t^2 \frac{(1+\nu)}{2\pi E} \left| \frac{1}{|\vec{r}|} \begin{pmatrix} 1 & 0 \\ 0 & \frac{\nu}{|\vec{r}|^3} \end{pmatrix} \frac{x^2}{xy} \frac{xy}{y^2} \right| \text{ for } |\vec{r}| \gg t, \text{ with } \vec{r} = \begin{pmatrix} x \\ y \end{pmatrix} \tag{Eq. S14}
\]

Here \(\nu\) is the Poisson’s ratio of the crystal’s elastic medium, and \(E\) is its Young’s modulus. The Fourier transform of Eq. S13 within the \(xz\)-plane yields the energy density of a single Fourier component \(\psi_{\vec{k}}\), which for sufficiently small \(k \equiv |\vec{k}| \ll t^{-1}\) reads

\[
 f_{\text{stripe}}(\vec{k}) \sim \frac{t}{2} \left[ \kappa k^2 - t \left| S^* \vec{k} \right|^2 \frac{1+\nu}{2\pi E k} \right] \left| \psi_{\vec{k}} \right|^2 \tag{Eq. S15}
\]

Here we have used the fact that \((\nabla \psi)_{-\vec{k}} = (\nabla \psi)^*_{\vec{k}} = -i \vec{k} \psi^*_{\vec{k}}\) for real-valued \(\psi(\vec{r})\), where \(^*\) denotes the complex conjugate. Eq. S15 encourages reinterpretation of the energetic competition in Eq. S12 through an effective “scale dependent surface tension” of the order parameter \(\kappa_{\text{eff}}(k) \equiv \kappa - t \left| S^* \right|^2 \frac{1+\nu}{2\pi E k}\). In this case Eq. S13 is minimized for a very particular value of \(k\) at equilibrium.
Here $\tilde{S}_{x,z}^*$ denotes the principle (maximal magnitude) component of $S^*$ projected onto the xz-plane, and $\Delta \alpha_{x,z}/\alpha_{x,z}$ denotes the change in unit cell size associated with the phase transition, projected along $\tilde{S}_{x,z}^*$. The orientation of $\vec{k}_{eq}$ is likewise expected to match that of $\tilde{S}_{x,z}^*$, provided that this principle component is sufficiently large compared with the other principle components. When this criterion is not satisfied, further minimization of elastic energy gain can be realized through emergence of more interesting equilibrium textures (51). However, as observed with nano-imaging in the present case of $\alpha$-(BEDT-TTF)$_2$I$_3$, this “principle” orientation is definitively the crystallographic a-axis.

This result accords with the length scale $\zeta$ indicated in the main text (Eq. 6) and follows closely derivations for the scale of phase inhomogeneities derived for other strain-coupled phase transitions in equilibrium (6). Physically speaking, Eq. S16 reflects the outcome of two competing real-space contributions to the system free energy. Long-range strain interactions (in proportion to $E$) tend to “thin” the length scale of inhomogeneities down to the scale of their depth $t$ so as to mitigate their elastic energy in a clamped environment, whereas short-range or microscopic domain-wall energetics tend to “coarsen” these inhomogeneities so as to reduce inter-phase interfacial energies. At the intrinsic equilibrium length scale $\zeta$, these effects precisely cancel. In principle, nano-scale sensitivity to $\zeta$ affords sensitivity to the energetics of both phenomena.

Note according to that Eq. S16, both the crystal thickness $t$ and the length scale $\zeta$ together quantitatively determine the periodicity of phase coexistence. If the thickness $t$ becomes too large compared to $\zeta$, it is expected that Eq. S14 no longer provides a quantitative description of the elasticity problem associated with domain boundaries. In this instance the details of the interplay between $t$, $\zeta$, and $k_{eq}$ become more subtle, and a more detailed formulation of the elasticity problem is needed. This scenario may be important to the nature of phase coexistence in much thicker crystals, and will be considered in forthcoming work.

While the phenomenology remains as stated above, the precise equality in Eq. S16 can be modified by a factor of order unity when considering that higher harmonics of $\vec{k}_{eq}$ arise in $\psi(\vec{r})$ to mutually minimize local terms in the Landau free energy (such as the temperature-dependent “double-well” potential $f_{CO}(\psi, T)$, not shown in Eq. S12). We therefore use a numerical approach to accurately calculate the equilibrium configuration of $\psi(\vec{r})$, which we describe in the following section.

\begin{equation}
\begin{aligned}
k_{eq} \sim & \frac{1}{2\pi} \frac{1+v}{E \kappa} |\tilde{S}_{x,z}^*|^2 \sim \frac{1}{2\pi(1+v)} \left( \frac{1-v}{1-2v} \right)^2 E \frac{\Delta \alpha_{x,z}}{\alpha_{x,z}} \equiv \left( \frac{t}{\zeta} \right) \cdot \frac{2\pi}{\zeta}
\end{aligned}
\end{equation}

(Eq. S16)
Minimization of Landau free energy.

In thermal equilibrium, the spatial configuration adopted by the order parameter function $\psi(\vec{r})$ is that which minimizes the Landau free energy $F$ presented in Eq. 1 of the main text. This configuration can be computed by solving the Euler-Lagrange differential equations, given the free energy density $f$ presented in Eq. S1

$$\frac{\partial f}{\partial \psi} - \nabla \cdot \frac{\partial f}{\partial \nabla \psi} = \mu(\vec{r}) = 0 \quad \text{for all } \vec{r} \text{ in } V \quad \text{(Eq. S17)}$$

Here $\mu(\vec{r})$ represents a local “chemical potential” for the system, expressions for which were derived accounting for the long-range strain energy contribution $f_L(\vec{r})$ of Eq. S10; these will be detailed in forthcoming publication. We implement an iterative solution to Eq. S10 based on a Cahn-Allen model for kinetics of a non-conserved order parameter

$$\frac{d\psi(\vec{r})}{dt} = -R \mu(\vec{r}) \quad \text{(Eq. S18)}$$

Here $R$ is a (ad hoc) positive rate constant, and time-stepping from an initial “trial” configuration $\psi_0(\vec{r})$ is accomplished by solving Eq. S18 via an implicit Crank-Nicholson scheme at incremental times $t$, using the finite element solver FEniCS evaluated at ever-increasing time-steps $\Delta t$ chosen to bring the system rapidly and arbitrarily close to the asymptotic condition $\mu(\vec{r}) = 0$. At this point the kinetics are concluded, resulting in an equilibrium configuration $\psi(\vec{r})$ that (though perhaps locally / metastably) minimizes the global Landau free energy $F$ at a particular temperature $T$.

Evaluation of $\psi(\vec{r})$ at each time step is performed on a mesh of size $66 \times 10 \mu m^2$ along x- and z-directions, respectively, with a total mesh size of $666 \times 100$. The “random field” contribution $D(\vec{r})$ appearing in Eq. S1 is generated from a gaussian distribution of random numbers with a small characteristic amplitude compared with amplitude of the temperature-dependent “double well” potential $f_{CO}(\psi, T)$. The presence of this small random field is sufficient to yield realistic “nucleation” of the charge-ordered phase during time evolution from the uniform initial phase.

For the results presented in Fig. 5A of the main text, calculations of equilibrium configurations of $\psi(\vec{r})$ were performed at a variety of temperatures ranging across $T=T_{CO}$. In this case, the initial configuration of $\psi(\vec{r})$ chosen for each temperature consisted of the equilibrium configuration obtained from the previous temperature. In this way, evolution of the charge-
ordered phase distribution across $T < T_{CO}$ to $T > T_{CO}$ could be predicted in a fashion following closely the actual experimental protocol, which consisted of stepwise changes in temperature across the transition.