

## Supporting Information

**Local Structural Disorder in FeCl<sub>3</sub>-SbCl<sub>5</sub>-Intercalated Graphite: Revealing the Disconnect Between Long-Range Periodicity and Nanoscale Intercalant Arrangement:**

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## **Supplementary Experimental Details**

This Supporting Information provides additional experimental details and analyses that complement the Experimental section in the main text.

### **TEM Image Acquisition**

Transmission electron microscopy (TEM) observations were carried out using an older-generation transmission electron microscope operated at an accelerating voltage of 200 kV. Images were recorded on photographic film and subsequently digitized by scanning. Because the intercalated graphite samples were highly susceptible to electron-beam damage, TEM observations were performed under low-dose conditions. As a consequence, the acquired images exhibit noticeable shot noise and relatively weak contrast.

### **Image Processing Procedure**

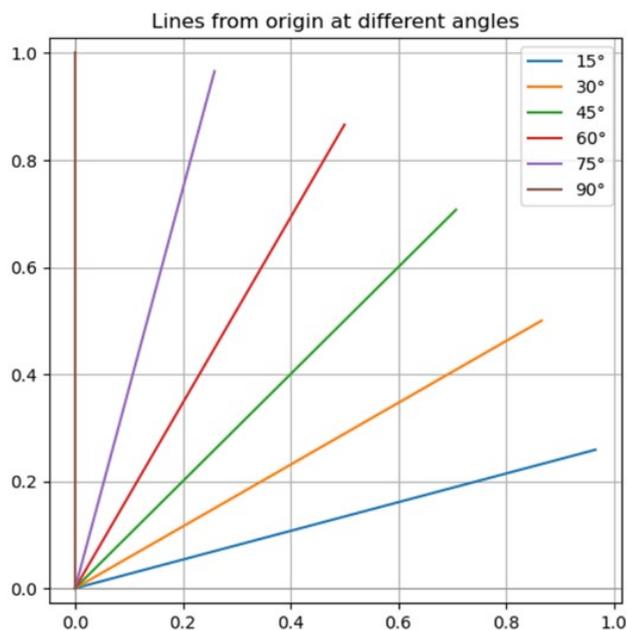
TEM image processing was performed using Fiji (ImageJ version 1.54p). To reduce shot noise while preserving structural information, a uniform median filter (radius = 2 pixels) followed by a uniform Gaussian blur ( $\sigma = 2$  pixels) was applied to the entire image. No local, selective, or feature-specific processing was performed.

The spatial calibration of the TEM images corresponds to 1 nm = 75 pixels, indicating that the applied filter size corresponds to approximately 0.027 nm. This length scale is significantly smaller than the characteristic lattice spacings of graphite and intercalated structures, ensuring that crystallographic or structural features were not altered by the image processing.

### **Camera Re-Imaging and Angular Analysis**

The obtained TEM image was re-imaged at different camera tilt angles to examine the visibility of lattice features as a function of viewing geometry. Photographs were taken using an iPhone 13 equipped with a 12 MP BSI-CMOS sensor.

During re-imaging, the camera tilt angles were varied systematically at 90°, 75°, 60°, 45°, 30°, and 15°, as illustrated in Fig. S1. The endpoints of the radial lines in Fig. S1 indicate the corresponding positions of the camera lens relative to the image plane.



3

Fig. S1 The lattice image was placed at the origin, and radial lines were drawn at 15°, 30°, 45°, 60°, 75° and 90°.

### Supplementary Discussion

The  $\text{FeCl}_3\text{-SbCl}_5$  intercalated species are expected to exhibit local structural variability within the graphite interlayers. Even when the contrast in the TEM images is not pronounced, peak broadening and irregularities observed in the X-ray diffraction profiles may manifest as local structural inhomogeneity in the TEM images.

These observations suggest that TEM and X-ray diffraction provide complementary structural information at different length scales, rather than redundant descriptions of the same structural features

### Quantitative Derivation of Molar Ratios from HR-TEM Stacking Sequences

To quantify the local chemical composition from HR-TEM images, we employed a layer-counting method combined with established in-plane density models for metal-chloride GICs.

The analyzed sequences represent typical localized regions captured via HR-TEM for each stage.

Sequence A (From Stage 3 based GIC):

G(FeCl<sub>3</sub>)G(SbCl<sub>5</sub>)GG(FeCl<sub>3</sub>)GG(SbCl<sub>5</sub>)GG(FeCl<sub>3</sub>)GGGG(FeCl<sub>3</sub>)GGG(FeCl<sub>3</sub>)GGG(FeCl<sub>3</sub>)G(SbCl<sub>5</sub>)G(FeCl<sub>3</sub>)GGG(FeCl<sub>3</sub>)GG(SbCl<sub>5</sub>)G

Sequence B (From Stage 5 based GIC):

G(FeCl<sub>3</sub>)GGGGG(FeCl<sub>3</sub>)GGGGGGGGGG(FeCl<sub>3</sub>)GGG(SbCl<sub>5</sub>)GG(FeCl<sub>3</sub>)GGG(SbCl<sub>5</sub>)GGG(SbCl<sub>5</sub>)GGG(FeCl<sub>3</sub>)GGGGG(FeCl<sub>3</sub>)GG(SbCl<sub>5</sub>)G

### 1. Structural Assumptions:

The molar density of each intercalant relative to the graphite layers was defined based on literature values [32]:

FeCl<sub>3</sub> layer: C<sub>6</sub> unit per molecule (FeCl<sub>3</sub> / 6C).

SbCl<sub>5</sub> layer: C<sub>12</sub> unit per molecule (SbCl<sub>5</sub> / 12C).

Calculation Unit: We define one graphite layer as 6 carbon atoms (C<sub>6</sub>) for consistent scaling.

### 2. Calculation Formula

The molar ratio is derived as follows:

$$C : n(\text{FeCl}_3) : n(\text{SbCl}_5) = (\text{NG} \times 6) : (\text{NF} \times 1) : (\text{NS} \times 0.5)$$

where NG, NF, NS are the number of observed layers for Graphite, FeCl<sub>3</sub> and SbCl<sub>5</sub> respectively.

### 3. Application to Observed Sequences:

For Stage 3 based GIC Sample (NG =26, NF =8, NS =4)

$$C = 26 \times 6 = 156$$

$$\text{FeCl}_3 = 8 \times 1 = 8$$

$$\text{SbCl}_5 = 4 \times 0.5 = 2$$

$$\text{Molar Ratio: } C_{156}(\text{FeCl}_3)_8(\text{SbCl}_5)_2 = C_{78}(\text{FeCl}_3)_4(\text{SbCl}_5)_1$$

For Stage 5 based Sample (NG =38, NF =6, NS =4)

$$C = 38 \times 6 = 228$$

$$\text{FeCl}_3 = 6 \times 1 = 6$$

$$\text{SbCl}_5 = 4 \times 0.5 = 2$$

$$\text{Molar Ratio: } C_{228}(\text{FeCl}_3)_6(\text{SbCl}_5)_2 = C_{114}(\text{FeCl}_3)_3(\text{SbCl}_5)_1$$