<u>Supporting Information</u>: Dielectrophoretic assembly of liquid-phaseexfoliated TiS₃ nanoribbons for photodetecting applications

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MATERIALS AND METHODS

Preparation of colloidal suspensions by liquid-phase exfoliation. 10 mL of isopropanol (IPA) were added to 10 mg of bulk TiS₃, and the heterogeneous mixture was stirred and sonicated for 1 min. The obtained dispersion was sonicated for 1 h in a Fisher Scientific FB 15051 ultrasonic bath (37 kHz, 280 W, ultrasonic peak max. 320 W, sine-wave modulation) thermostated at 20 °C. 750 μ L of the resulting suspension (suspension A) were kept for further experiments and characterization. The rest of the suspension was centrifuged (990 g, 30 min, 25 °C, Beckman Coulter Allegra® X-15R, FX6100 rotor, radius 9.8 cm). The resulting sediment was redispersed (30 s sonication) in 10 mL *i*PrOH (suspension B), while the supernatant (including partially dispersed light sediment) was subjected to a second round of centrifugation (990 g, 30 min, 25 °C). The supernatant was discarded and the sediment was redispersed in 1 mL *i*PrOH (suspension D). 750 μ L of suspension B were kept for further studies and the rest of the suspension was centrifuged at lower speed (30 g, 30 min, 25 °C). ~8 mL of the supernatant were carefully isolated (suspension C). The remaining supernatant and sediment were discarded.

Scanning Electron Microscopy. Samples and devices were observed with a Auriga FE-SEM microscope operated at 3 kV in Secondary Electron (SE) mode (in-lens for Figure 1a of the main text; conventional Everhart-Thornley for all other figures).

Raman spectroscopy. Samples were analyzed with a Bruker Senterra confocal Raman microscopy instrument (Bruker Optik, Ettlingen, Germany) under the following conditions: objective NA 0.75, 50×; laser excitation: 532 nm, 2 mW. As-synthesized bulk TiS₃ was pressed on a glass slide and the data shown correspond to typical

spectra acquired in different regions of the solid sample. Colloidal suspensions were dried on glass slides at 40 °C and their respective spectra result from the average of 10 measurements performed over their surface.

UV-Vis-NIR spectroscopy. Colloidal suspensions were diluted in IPA and their extinction spectrum (both absorption and scattering are measured using such a piece of equipment) was recorded with a Cary 5000 spectrophotometer, Agilent Technologies (wavelength range: 175-3300 nm).

Device fabrication. The parallel interdigitated electrodes were fabricated on a silicon (Si) wafer covered with a 300 nm CVD-grown silicon oxide (SiO₂) layer. The interdigitated pattern was developed on the Si/SiO₂ substrate via a standard photolithography process. A 10 nm chromium layer (adhesion material) and a 100 nm gold layer (electrode material) were deposited onto the developed surface by thermal evaporation. Lift-off was carried out for the gold layer through wet etching of the unexposed surface. The channel gap between the parallel electrodes is 5 μm.

Dielectrophoresis. The DEP process was conducted over the interdigitated electrodes structures. The set-up consists in a function generator used to create an oscillating electric field between the electrodes and in an oscilloscope used to monitor the voltage change in real time during the DEP process. The frequency and the peak-to-peak voltage of the AC signal are 1 MHz and 10 V, respectively. As for the deposition and horizontal alignment of TiS₃, a droplet of suspended TiS₃ nanoribbons was placed onto the electrode gaps in the presence of the non-uniform electric field. The applied electric field polarizes the nanoribbons, thus creating dipoles that align along the electric field lines.



SEM ANALYSIS OF BULK TIS₃

Figure S1: Low-magnification SEM images of bulk TiS₃. a) Magnification ×543. b) Magnification ×232.



Figure S2: Raman spectra of liquid-phase-exfoliated TiS_3 (average of 10 measurements and normalized at 368 cm⁻¹). a) Suspension A. b) Suspension B. c) Suspension C. d) Raman spectra of all colloidal suspensions normalized at 368 cm⁻¹.

 TiS_3 bands at 100 cm⁻¹ (weak, B_g), 174 cm⁻¹ (strong, A_g), 256 cm⁻¹ (shoulder, B_g), 273 cm⁻¹ (shoulder, A_g), 298 cm⁻¹ (strong, A_g), 368 cm⁻¹ (strong, A_g), 556 cm⁻¹ (strong, A_g), 641 cm⁻¹ (broad, 273 cm⁻¹ + 368 cm⁻¹).

Extra bands at 151 cm⁻¹ (strong, E_g), 201 cm⁻¹ (weak, E_g), 400 cm⁻¹ (weak, B_{1g}), 515 cm⁻¹ (weak, B_{1g} and A_{1g}) and at 641 cm⁻¹ (weak, superimposed to TiS₃, E_g) indicate the presence of TiO₂



Figure S3: Typical Raman spectra (normalized at 368 cm⁻¹) obtained from the analysis of various sites of bulk TiS₃.



Figure S4: Typical Raman spectra obtained from the analysis of various devices built from liquid-phase exfoliated TiS_3 . The strong peak at 520 cm⁻¹ is a Raman peak of the crystalline silicon substrate.



Figure S5: Extinction spectra of liquid-phase-exfoliated TiS_3 . a) Suspension A. b) Suspension B. c) Suspension C. d) Extinction spectra of all suspensions normalized at 250 nm.



Figure S6. Tauc plot drawn from the absorption data of suspension D. The optical bandgap energy corresponds to the point where the linear fit (red line) meets the X axis.

The absorption studies were limited to wavelengths below 1180 nm due to the strong absorption of IPA that prevents any measurement in the far infrared. Additionally, as the nanoribbons have micrometric length, non-

negligible scattering is observed and the resulting spectra must be considered as extinction spectra (that is, the sum of absorption and scattering spectra).



DIELECTROPHORESIS-BASED LIQUID-PHASE-EXFOLIATED SOLID STATE DEVICES

Figure S7: Analysis of the angular distribution of TiS₃ nanoribbons between gold electrodes from the device discussed in the main text.



Figure S8: a) Length distribution of the assembled TiS_3 nanoribbons. The length was measured from 123 nanoribbons identified in the SEM image of Fig. S6. b) Angular distribution of the assembled TiS_3 nanoribbons where 0° correspond to a nanoribbon oriented along the source-drain electrodes direction.



Figure S9: SEM images of the devices made from colloidal suspensions of exfoliated TiS₃. a-b) Made from suspension A. c-d) Made from suspension B. e-f) Made from suspension C.



Figure S10: Time response of a device made from suspension A upon modulated optical excitation with a light wavelength of 405 nm.



Figure S11: a) Time response of the main text device upon modulated optical excitation with a light wavelength of 638 nm and increasing laser powers up to 47 mW. The measurements are carried out at a bias voltage of 10 mV. b) Time response at two different excitation wavelengths, λ = 405 nm circular spot diameter 120 µm, λ = 638 nm rectangular spot dimensions 100x45 µm. The optical power density of 4 W/mm² is the same for both illuminations. The time axis of each dataset has been normalized by the time constant (10%-90% rule) of the curve.