BEDT-TTF (98%, Alfa Aesar, USA) was used as received. Platinum foils (99.99%) were used after being polished with fine sand paper and cleaned in an ultrasonic bath of acetone for 20 min, 0.1 M HCl for 20 min, deionized water for 20 min, and ethanol for 20 min. The products were characterized and analyzed by scanning electron microscopy (SEM, JEOL JSM-6700F), transmission electron microscopy (TEM, JEOL 2010). For X-Ray diffraction (XRD) and X-Ray photoelectron spectrometer (XPS) characterizations the films were used without substrates. The XRD patterns were recorded with a Japan Rigaku D/max-2500 rotation anode X-ray diffractometer equipped with graphite monochromatized Cu Kα radiation (λ=1.5418Å), in the 2θ range from 5° to 60°. The XPS was collected on VG Scientific
ESCALab220i-XL X-Ray photoelectron spectrometer, using Al Kα radiation as the excitation sources. The banding energies obtained in the XPS analysis were corrected with reference to C 1s (284.8eV).

The Fourier transform infrared (FT-IR) spectra were recorded on a Bruker EQUINOX55 Fourier transform infrared spectrometer using KBr pellet technique in the spectra range of 4000-500cm⁻¹.

The field emission measurements were carried out in a vacuum chamber of 5 × 10⁻⁷ Pa at room temperature under a two-parallel-plate configuration. The platinum foil with (BEDT-TTF)$_2$Cu(SCN)$_2$ was stuck onto a stainless-steel sample stage using conducting glue as the cathode. Another parallel stainless-steel plate served as the anode. The samples’ areas were about 0.061 cm² for the (BEDT-TTF)$_2$Cu(SCN)$_2$ nanorods grown on the Pt foils under 100µA/cm² and 0.062 cm² for the (BEDT-TTF)$_2$Cu(SCN)$_2$ nanorods grown on the Pt foils under 200µA/cm². The distance between the anode and cathode is 100 µm for the (BEDT-TTF)$_2$Cu(SCN)$_2$ nanorods grown on the Pt foils under 100µA/cm² and 300 µm for the (BEDT-TTF)$_2$Cu(SCN)$_2$ nanorods grown on the Pt foils under 200µA/cm². A voltage with a sweep step of 50 V was applied between the anode and cathode to supply an electric field.

1. Experimental Section

A platinum foil (cathode) and an ITO glass (anode) were used as the working electrodes. A drawing of a typical electrocrystallization cell was described in Figure S1. 3.5mg (0.09mmol) BEDT-TTF, 8mg (0.07 mmol) CuSCN, 18mg (0.19 mmol) KSCN, and 8mg (0.03 mmol) 18-Crown-6 were dissolved in10ml of 1,1,2-trichloroethane (TCE) and stirred vigorous for 3 h at 50 °C. The mixture was cooled down to 30 °C, and the remainder of undissolved CuSCN and KSCN was precipitated on the bottom of the electrocrystallization cell. (BEDT-TTF)$_2$Cu(SCN)$_2$ was completely grown on the surface of the anode ITO glass after reaction for 4h at 30 °C under a constant current density of about 100µA/cm² and 200µA/cm², respectively. The (BEDT-TTF)$_2$Cu(SCN)$_2$ nanorods can be also obtained on the surface of Pt foil, if the anode ITO glass was replaced with Pt foil. The morphology structure and compositions of the products were investigated by SEM, TEM, EDS, FT-IR, XRD, and XPS.
**Figure S1.** An electrocrystallization cell used in the experiments.

2. The reaction scheme of the \((\text{BEDT-TTF})_2\text{Cu(SCN)}_2\).\(^{1-2}\)

\[
\begin{align*}
\text{BEDT-TTF} & \quad \overset{-e}{\longrightarrow} \quad \text{BEDT-TTF}^+ \\
\text{KSCN} + \text{CuSCN} + C_{12}H_{24}O_6 & \quad \longrightarrow \quad (K^+ C_{12}H_{24}O_6)[\text{Cu(SCN)}_2]^+ \\
\text{BEDT-TTF} + \text{BEDT-TTF}^+ + [\text{Cu(SCN)}_2]^+ & \quad \longrightarrow \quad (\text{BEDT-TTF})_2\text{Cu(SCN)}_2
\end{align*}
\]

**Scheme S1.** Scheme of the reaction process of the \((\text{BEDT-TTF})_2\text{Cu(SCN)}_2\).

**Figure S2.** Stacking mode of the \(\kappa-(\text{BEDT-TTF})_2\text{Cu(SCN)}_2\) molecules.

3. SEM images of \((\text{BEDT-TTF})_2\text{Cu(SCN)}_2\).
Figure S3a. (A) SEM images of the (BEDT-TTF)$_2$Cu(SCN)$_2$ grown on the Pt foils under 5µA/cm$^2$, (B) 50µA/cm$^2$, (C) 300µA/cm$^2$, (D) 400µA/cm$^2$, and (E) 500µA/cm$^2$.

Figure S3a. (A) SEM images of the (BEDT-TTF)$_2$Cu(SCN)$_2$ grown on the ITO under 50µA/cm$^2$, (B) 150µA/cm$^2$, (C) 250µA/cm$^2$, (D) 300µA/cm$^2$.

4. Energy-Depressive Spectrum (EDS) of (BEDT-TTF)$_2$Cu(SCN)$_2$ nanorods.
For energy-depressive X-microanalysis, the (BEDT-TTF)$_2$Cu(SCN)$_2$ nanorods grown on the Pt foils were used directly. The results of energy-depressive spectrum (EDS) showed that the (BEDT-TTF)$_2$Cu(SCN)$_2$ nanorods grown on the Pt foils were composed of copper, carbon, nitrogen and sulphur elements.

5. XPS of (BEDT-TTF)$_2$Cu(SCN)$_2$ nanorods.

Figure S4. (A) EDS of the (BEDT-TTF)$_2$Cu(SCN)$_2$ nanorods grown on the Pt foils under 100μA/cm$^2$. (B) EDS of the (BEDT-TTF)$_2$Cu(SCN)$_2$ nanorods grown on the Pt foils under 200μA/cm$^2$.

Figure S5. XPS data in the Cu2p3/2 and 2p1/2 (A), N1s (B), C1s(C), and S2p (D) for the (BEDT-
TTF)$_2$Cu(SCN)$_2$ nanorods grown on the Pt foils.

The success of the preparing the (BEDT-TTF)$_2$Cu(SCN)$_2$ was further confirmed by the XPS spectra. The 2p$_{1/2}$ and 2p$_{3/2}$ signal (932.5 eV and 952.0 eV) exhibit essentially identical binding energies for Cu2p orbital in accord with Cu (I). The N1s orbital appears as a single feature at 398.4 eV. Also, the C1s orbital appears as a single feature at 284.8 eV and the S2p orbital appears as a single feature at 163.7 eV. All these consist with the (BEDT-TTF)$_2$Cu(SCN)$_2$.

Reference:
