Electronic Supplementary Material

Tuning Reaction Processes for the Synthesis of Micron and Nanometer Sized, Single Crystalline Lamellae of Copper 7,7,8,8-tetracyano-*p*-quinodimethane (Phase II) with Large Area

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In order to identify w diffusion of Cu^{2+} through the barrier of the Au layer, Au film slides were prepared and soaked in $CuSO_4$ solution for 6 h and then rinsed five times by pure water to wash away the physically adsorbed Cu^{2+} on the surface of the Au slides, and then characterized by XPS. The results are shown in Fig. S-1; the obvious satellite peaks of Cu^{2+} demonstrate the existence of a large amount of Cu^{2+} in the Au films, i.e., the diffusion of Cu^{2+} into the Au films.

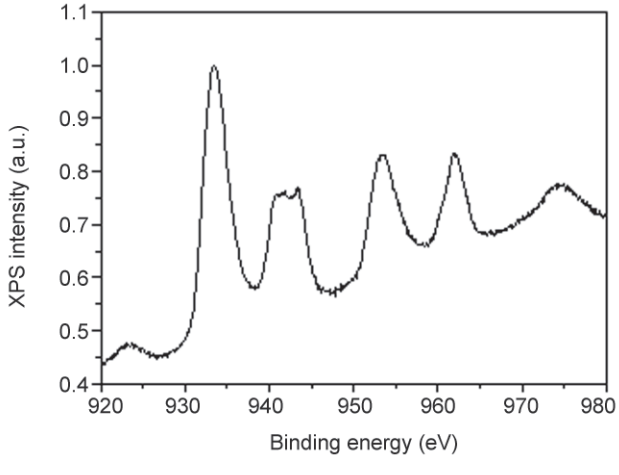

Figure S-1 XPS of Au films soaked in Cu^{2+} solution clearly demonstrating the existence of Cu^{2+} in the films

The mixture of the lamellar structures (phase II) with rod-like (Fig. S-2(a)) or other (Figs. S-2(b)–(d)) phase I morphologies demonstrated that both phases were obtained in thin films of CuTCNQ when either the "displacement reaction" was eliminated or the thickness of Au barrier was reduced from 100 nm down to 20–30 nm, thus weakening the control of the diffusion processes.

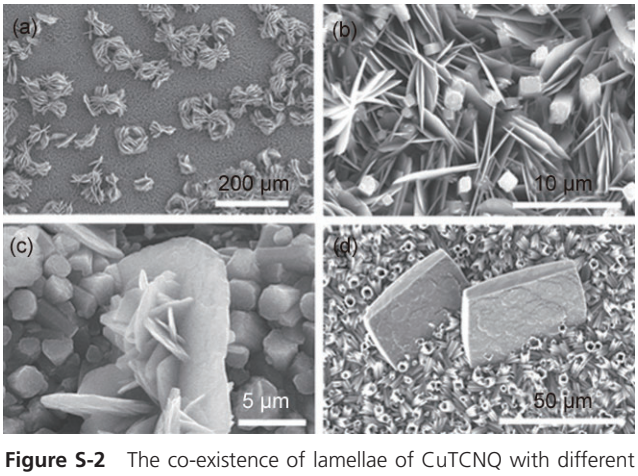

Figure S-2 The co-existence of lamellae of CuTCNQ with different kinetic phase I products: (a) rods, (b) square columns, (c) polygonal columns, (d) tubes. The rods, square columns, polygonal columns and tubes are all phase I materials (see Refs. [39] and [41] in the main text)

Our preliminary results using an individual lamella indeed exhibited switching phenomenon (Fig. S-3), indicating the potential application of CuTCNQ in memory and switching elements. The devices were fabricated by depositing electrodes on a SiO_2 substrate, then transferring a lamella across the electrodes, and finally vacuum-depositing the gap electrodes on the crystal again. The switching behavior could be observed with (i) different metals as the electrodes, and (ii) with symmetrical or asymmetrical device configurations. Detailed investigations are underway in our laboratory.

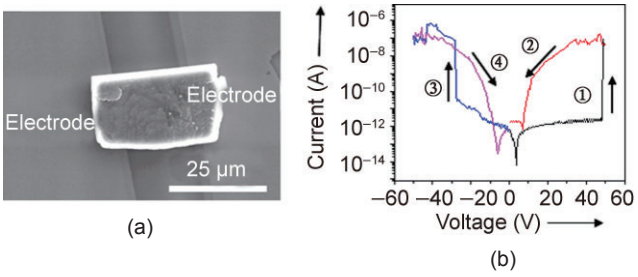

Figure S-3 SEM image (a) and current–voltage characteristics (b) of a device based on an individual lamella