# Interfacing Solution-Grown $C_{60}$ and (3-pyrrolinium)($CdCl_3$) Single-Crystals for High-Mobility Transistor-Based Memory Devices

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**Abstract:** Organic field effect transistors (FETs) based on organic single-crystals are ideal candidates for high-performance transistor-based memory devices due to their high charge mobility; however, they have not been largely considered for memory devices due to the practical difficulty in interfacing organic single-crystals with memory functional materials such as ferroelectrics. Here, we demonstrate that well-aligned ferroelectric single-crystals of (3-pyrrolinium)($CdCl_3$) can be prepared, from solution, on top of well-aligned semiconducting $C_{60}$ single-crystals, using an orthogonal solvent. By showing a large memory window of 66 ± 7 V as well as a high electron mobility of 1.28 ± 0.41 $cm^2V^{-1}s^{-1}$, these bilayered single-crystals are potentially useful for high-performance FET memory devices with high operation speed.

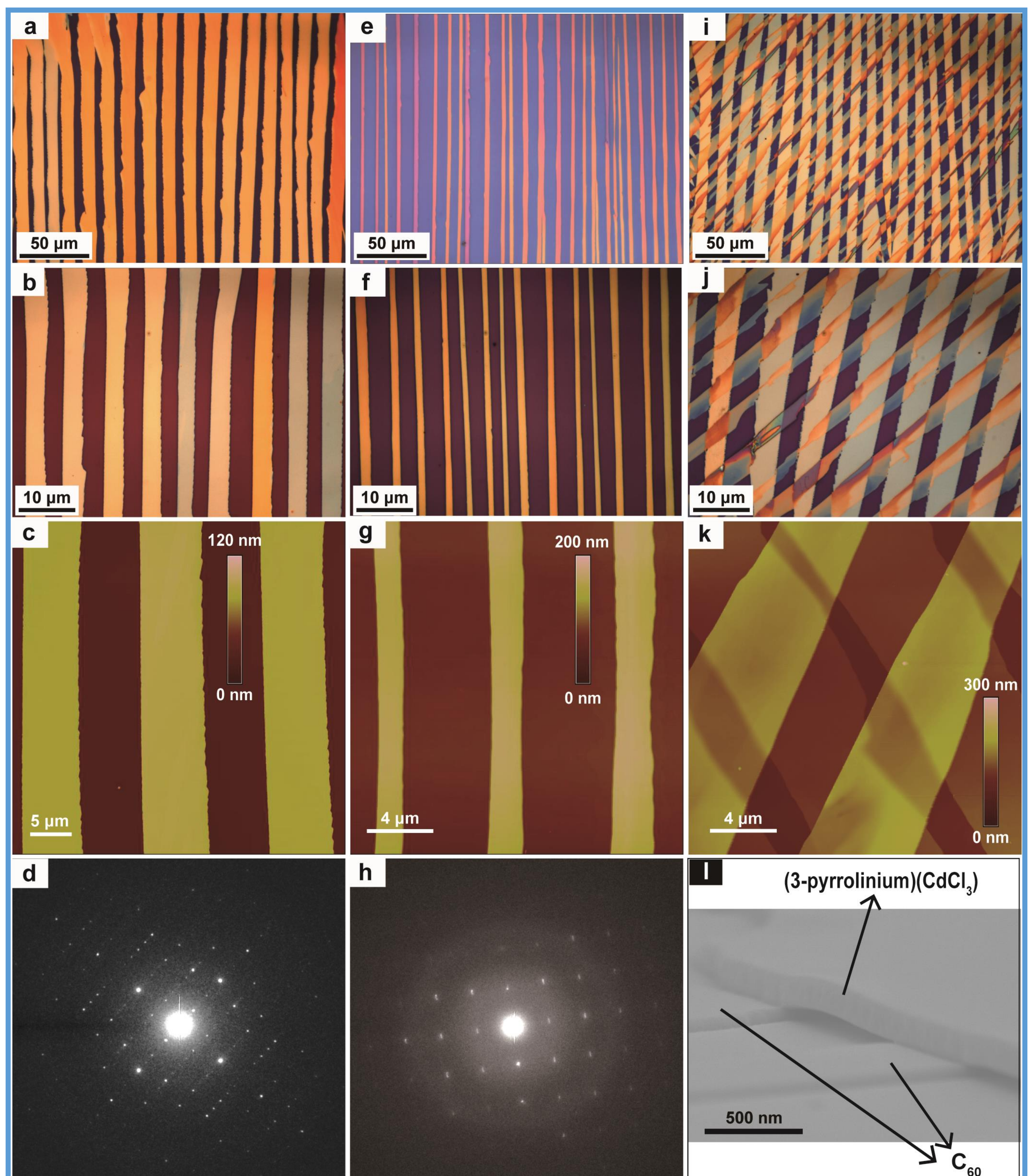


**Figure 1.** The morphologies and crystalline structures of $C_{60}$ crystals (a-d), (3-pyrrolinium)($CdCl_3$) crystals (e-h) and their bilayered heterojunctions (i-l), respectively. (a, b, e, f, i, j) Optical microscopy (OM) images; (c, g, k) AFM images; (d, h) SAED patterns showing single sets of the diffraction spots. (i) A scanning electron microscope (SEM) image (side view) of the bilayered heterojunctions.

In summary, aligned ferroelectric single-crystals of (3-pyrrolinium)($CdCl_3$) were grown, from solutions, onto aligned semiconducting $C_{60}$ single-crystals using an orthogonal solvent. Interfacing the molecular ferroelectric single-crystals with organic semiconducting single-crystals through the solution growth method provides a facile approach to fabricate high-performance FET-based memory devices. Expanding the material systems to construct varied multi-layered highly crystalline films should further help realization of multi-functional FETs based on organic single-crystals.

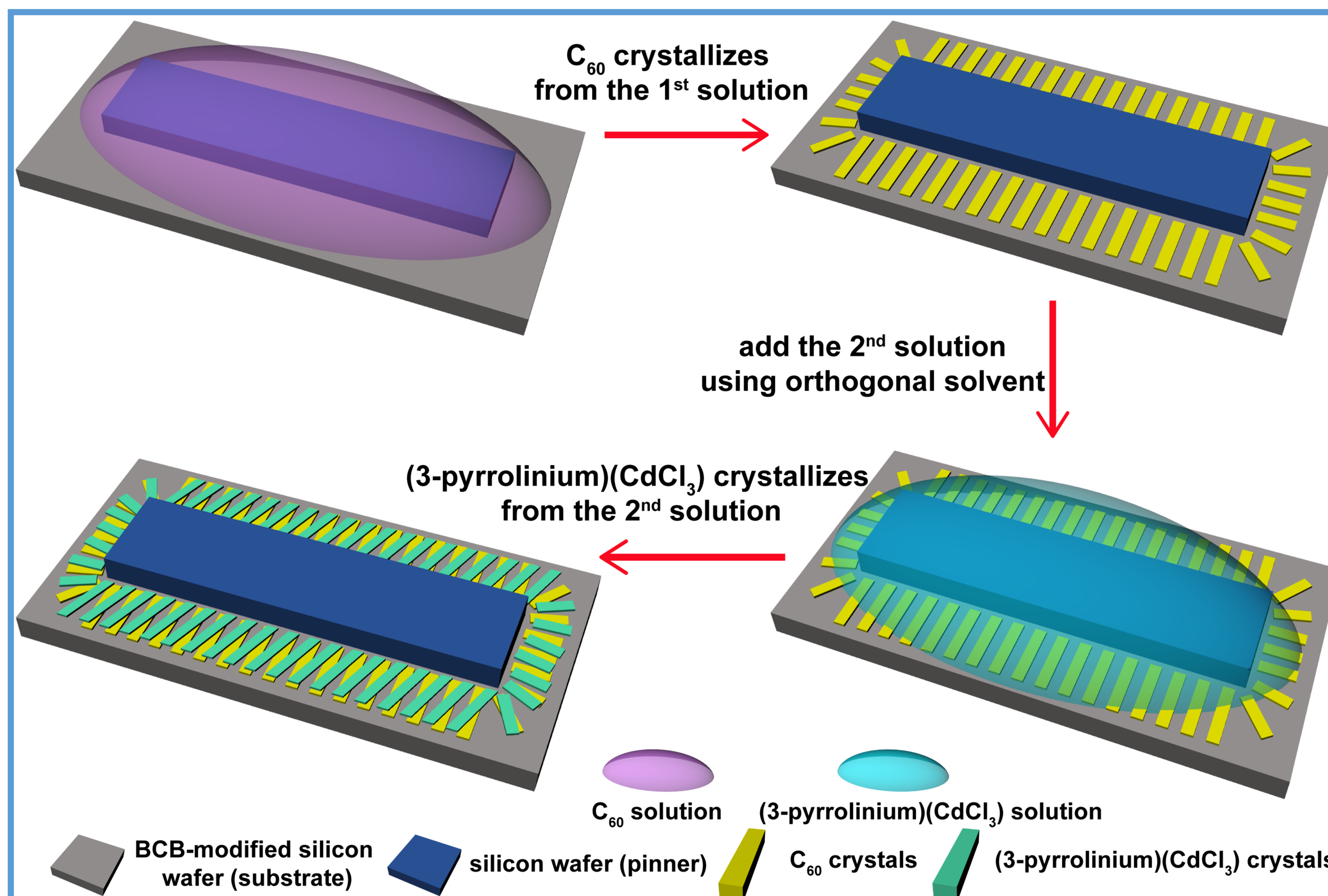


**Figure 2.** Schematic representations of the two-step crystallization process. A $C_{60}$ solution is first dropped on a BCB-modified silicon wafer with a smaller piece of wafer to pin the droplet. As the solvent evaporates and the droplet recedes slowly, $C_{60}$ molecules crystallize along the receding direction of the droplet. Subsequently, adding another droplet of (3-pyrrolinium)($CdCl_3$) solution in an orthogonal solvent results in bilayered crystals after the (3-pyrrolinium)($CdCl_3$) crystals form on top of the $C_{60}$ crystals. Crystallization in the top layer is affected by the morphology of the bottom layer and the orientation of the crystals is determined by the receding direction of the droplet as well as the surface topology. As a result, the crystal orientations in the top and bottom layers are not identical.

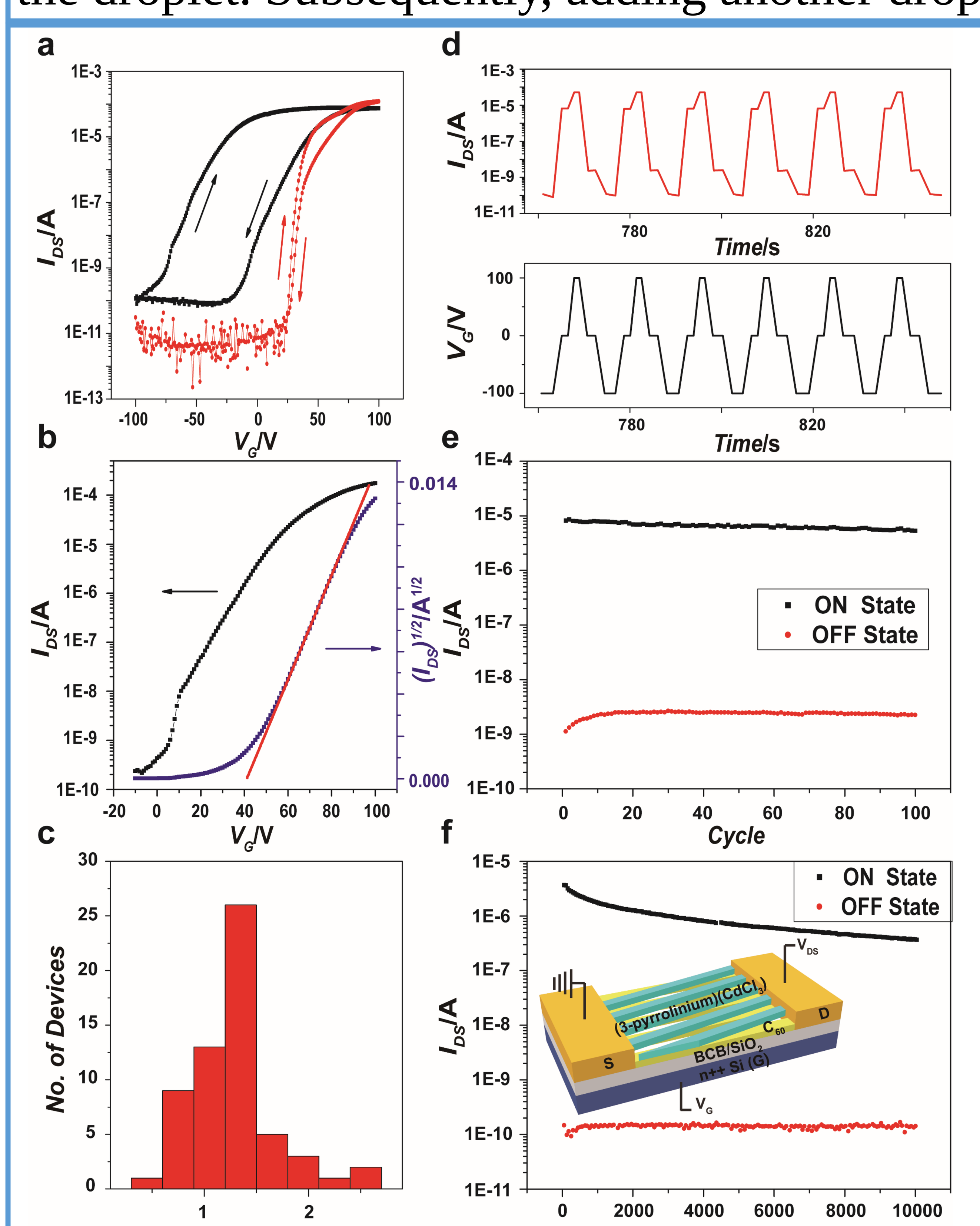


**Figure 3.** Charge transport characteristics of the (3-pyrrolinium)($CdCl_3$)/$C_{60}$ bilayered single-crystals. (a) Hysteresis characteristics of FETs based on $C_{60}$ crystals with (black dot) and without (red dot) a layer of (3-pyrrolinium)($CdCl_3$) crystals on top. (b) Typical transfer characteristics of the devices. (c) Histogram of electron mobility. (d, e, f) WRER cycles, endurance characteristics and retention time test of the memory devices, respectively. The inset in f is a schematic diagram of the FET configuration, where S is the source, D the drain and G the gate.

## References

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2. H. Y. Ye et al. Angew. Chem. Int. Ed. **2014**, 53, 11242.