Solution-processed, low cost thin films of layered semiconductors such as transition metal dichalcogenides (TMDs) are potential candidates for future printed electronics. Here, n-type electrolyte-gated transistors (EGTs) based on porous WS$_2$ nanosheet networks as the semiconductor are demonstrated. The WS$_2$ nanosheets are liquid phase exfoliated to form aqueous/surfactant stabilized inks, and deposited at low temperatures (T < 120 °C) in ambient atmosphere by airbrushing. No solvent exchange, further additives, or complicated processing steps are required. While the EGTs are primarily n-type (electron accumulation), some hole transport is also observable. The EGTs show current modulations > 10$^4$ with low hysteresis, channel width-normalized on-conductances of up to 0.27 $\mu$S $\mu$m$^{-1}$ and estimated electron mobilities around 0.01 cm$^2$ V$^{-1}$ s$^{-1}$. In addition, the WS$_2$ nanosheet networks exhibit relatively high volumetric capacitance values of 30 F cm$^{-3}$. Charge transport within the network depends significantly on the applied lateral electric field and is thermally activated, which supports the notion that hopping between nanosheets is a major limiting factor for these networks and their future application.

1. Introduction

Layered materials such as graphene, transition metal dichalcogenides (TMDs), black phosphorous etc. are intensely investigated for use in a broad range of applications in electronics, optoelectronics, and electrochemistry.[1–4] The past decade has seen numerous studies focused on the characterization of charge transport in these semiconductors in field-effect transistors (FETs), with particular emphasis on the influence of layer number,[9] contact/dielectric engineering,[6,7] and optoelectronic behavior at the monolayer limit.[8-9] Some key attributes driving interest in this field include atomically flat basal plane surfaces without dangling bonds, the possession of a variety of bandgaps (depending on the elemental composition and layer number),[10] good room temperature mobilities,[11,12] and the potential to produce heterostructures by combining sheets laterally or vertically.[13] These features, among others, make layered semiconductors attractive candidates for future-generation electronics either by enabling further channel scaling, new functionalities, or integration into transparent, flexible, and wearable devices.[1,14]

In addition to approaches that focus on the manipulation of individual layers, there is increasing interest in using scalable printing technologies to process layered semiconductors at low temperatures from inks into networks and thin films.[15] A major objective is to combine these films to form devices with reasonable performance at very low cost.[16] Torrisi et al. made an early step in this direction with ink-jet printing of conductive graphene sheet networks, which were also gated electrostatically to alter the network charge carrier density.[17] Since then, significant progress has been made regarding the understanding of processing layered materials into inks in large quantities[18] and controlling the mean flake geometry using size-selection techniques,[19] both of which are essential steps towards functional printing of these materials.[20,21] By now a number of groups have demonstrated solution-processed devices such as photodetectors,[15,22,23] chemical sensors,[24] and thin film transistors,[25,26] featuring layered semiconductors, especially members of the TMD family,[27] and printed electrodes and interconnects using graphene.[28] Recently, Kelly et al. deposited and characterized porous nanosheet networks of different TMDs by airbrush spraying.[29] In particular, they incorporated WSe$_2$ with other printed nanosheet network structures (composed of graphene and hexagonal boron nitride (h-BN)) to form all-layered p-type transistors that were gated with an ionic liquid (IL).[30] Similar to conjugated polymer and quantum dot (QD) based thin films, the porous nature of such nanosheet networks facilitates ion penetration into the internal network structure and thus
efficient gating of the whole film.\textsuperscript{[29,30]} Despite these recent developments, it is clear that the performance of printed semiconductor networks needs to be improved substantially for them to become competitive with more established printable material classes, especially conjugated polymers,\textsuperscript{[31]} single-walled carbon nanotubes (SWNTs),\textsuperscript{[32]} and solution-processed metal oxides.\textsuperscript{[33]} So far little is known about the processing requirements and ultimate device characteristics that could be achieved with TMD nanosheet films that promise low cost and scalable production. Importantly, for future printed, low-power complementary circuits both p-type and n-type transistors are required with equal carrier mobilities. Unlike single flake devices based on WS\textsubscript{2}, which show n-type or ambipolar transport,\textsuperscript{[34,35]} the demonstrated WS\textsubscript{2} nanosheet film transistors up to now only show hole transport.\textsuperscript{[26]}

With this study, we demonstrate the first n-type transistors based on liquid phase exfoliated layered WS\textsubscript{2} networks and characterize their electrical properties when gated using an ionic liquid. The nanosheet films are deposited at low temperatures from aqueous/surfactant-based inks without other additives and show substantially improved performance compared to previous efforts, allowing us to study their charge transport properties in more detail.

2. Results and Discussion

Figure 1a presents a schematic of the electrolyte-gated transistors (EGTs) prepared in this study. The channel consists of an electrolyte-permeable network of liquid phase exfoliated layered WS\textsubscript{2} nanosheets that was deposited from an aqueous/surfactant (sodium cholate) stabilized ink by airbrush deposition onto interdigitated source/drain electrodes (Figure 1b). The surface morphology of the deposited film was examined by scanning electron microscopy (SEM), revealing a porous, disordered network structure consisting of predominantly multilayered nanosheets with lateral dimensions of hundreds of nanometers (Figure 1c). These geometric features are consistent with values obtained from analysis of extinction spectra using established flake geometry metrics,\textsuperscript{[36]} with mean flake length of $<L> = 415$ nm and mean layer number of $<N> = 20$–30 (see Figure S1 in the Supporting Information).

To complete the devices, an ionic liquid (anhydrous and degassed) commonly used for EGTs (1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide, [EMIM][TFSI])\textsuperscript{[37,38]} contained by a polydimethylsiloxane (PDMS) frame was placed on the channel area and the large side-gate electrode. The addition of the electrolyte and subsequent electrical measurements were performed in dry nitrogen. The device characteristics were investigated through control of the gate voltage ($V_g$) and drain voltage ($V_d$) while the source was grounded (Figure 1a).

Current–voltage characteristics for a typical well-operating device are shown in Figure 2 (channel width, $W = 10$ mm, channel length, $L = 15$ mm). The transfer curves demonstrate the prominent n-type behavior (i.e., electron accumulation and transport). The gate leakage current ($I_g$) is fairly typical for electrolyte-gated thin films ($≈10^{-7}$ A)\textsuperscript{[37]} and the drain current ($I_d$) modulation was $>10^4$, compared with $≈500$ for the printed p-type WS\textsubscript{2} nanosheet network transistors recently reported by Kelly et al.\textsuperscript{[26]}

Transistors prepared by Kelly et al. with WS\textsubscript{2} networks only displayed p-type switching, a difference which may be associated with the ink preparation (i.e., liquid phase exfoliation in N-methyl-2-pyrrolidone and solvent transfer to isopropanol)\textsuperscript{[26]} For our devices, we found it important to soak the thin films in deionized (DI) water to remove the residual surfactant, followed by heating in nitrogen atmosphere ($T = 100$ °C). While soaking seems to be reasonably effective (Figure S2, Supporting Information), it is likely that some adsorbed cholate remains on the nanosheet surfaces. At present, little is known about the influence of such residues on transport in nanosheet networks and the extent to which they can or should be removed. However, this work emphasizes the need to systematically study the effect of adsorbates, and moreover, exploit them for tunable switching behavior. A number of studies have investigated this aspect of device processing,\textsuperscript{[39,40]} which is particularly interesting for networks deposited from solution as the environment could be readily varied homogeneously prior to deposition.

From the obtained current voltage characteristics of our WS\textsubscript{2} nanosheet EGT in Figure 2 we calculated channel width normalized on-conductances (defined as $G_{on} = I_{on}/V_d$ with $I_{on}$ at maximum $V_d$) of $G_{on}/W = 0.27$ and $0.16$ $\mu$S $\mu$m$^{-1}$ at drain biases of $V_d = 0.1$ V and $V_d = 1$ V, respectively. The on-conductance provides a straightforward measure of how readily the device can drive current between the source and drain. It is arguably a more useful metric for comparing different thin film transistors than the field-effect mobility, $\mu$. As recently emphasized,\textsuperscript{[41]} $\mu$ is susceptible to inaccurate reporting due to the additional and sometimes unknown parameters required for its calculation. In the case of EGTs the correct determination of the carrier density via the capacitance is particularly difficult. Compared to other classes of electrolyte-gated and printed semiconductors (see comparison of $G_{on}/W$ for a range of EGTs in

![Figure 1](image-url)
ohmic contacts are formed between the WS$_2$ network and the Au electrodes (see also Figure S4 in the Supporting Information). At present, it is unclear what the individual multilayer WS$_2$ flakes by Braga et al.\cite{34} The high effective capacitance achieved by electrolyte gating with an ionic liquid facilitates charge injection at the metal/semiconductor interface and reduces contact resistance for both holes and electrons.\cite{44} It also promotes effective trap filling in a network with a high density of structural defects (primarily at the flake edges). In addition to the trap state density, the observed threshold voltages in EGTs depend on the bandgap of the semiconductor. Among the common transition metal dichalcogenides, WS$_2$ possesses the largest bandgap.\cite{45} Therefore, other layered semiconductors such as the molybdenum or tungsten selenides and tellurides will likely show different threshold voltages under similar processing conditions.

Figure S3 and Table S1 in the Supporting Information) such as conjugated polymers\cite{29} and SWNTs,\cite{42,43} the WS$_2$ nanosheet networks still require substantial improvement. However, given that our understanding of the processing requirements and properties of printed nanosheet network semiconductors is still in its infancy, the achieved on-conductances are promising.

To gain insights into the relative influence of contact and channel resistances, we prepared devices with a range of channel lengths ($L = 2–35$ $\mu$m) at constant channel width ($W = 10$ $\mu$m). Such a sample set was deposited during a single airbrush deposition run to enable consistent film thickness (Figure S8, Supporting Information) and network morphology. As shown in Figure S7 in the Supporting Information) was not strongly dependent on the gate voltage sweep rate ($dV_g/dt$), in contrast to other reports for electrolyte-gated thin films,\cite{46} and is probably associated with trap states.\cite{47}

The linear, $\mu_{lin}$, and saturation, $\mu_{sat}$, mobilities were calculated using Equations (1) and (2), respectively:\cite{41}

$$\mu_{lin} = \frac{L}{t_{th} W C V_{th}} \left( \frac{\partial I_d}{\partial V_g} \right)$$

$$\mu_{sat} = \frac{2 L}{t_{th} W C V_{th}^2} \left( \frac{\partial I_d}{\partial V_g} \right)^2$$

Or likewise in their integrated forms

$$I_d = \frac{\mu_{lin} \cdot t_{th} W C V_{th} (V_g - V_{th}) \cdot V_d}{L}$$

Figure 2. a) Transfer and output characteristics for b) electron and c) hole accumulation conditions. The sweep rate for the transfer curves was 25 mV s$^{-1}$. The arrows indicate the sweep direction.

- $I_g (V_d = 1.0 \text{ V})$
- $I_g (V_d = 0.1 \text{ V})$
- $I_d (V_d = 1.0 \text{ V})$
- $I_d (V_d = 0.1 \text{ V})$

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that charge accumulation occurs throughout the volume of the channel thickness, \( t_{ch} \) was measured for each sample by contact profilometry; this modification is needed, as we expect that the current response is stable (Figure S13b, Supporting Information). The uncertainty in the thickness and precise area of the porous film increases the error for the estimated carrier mobility, although by less than 20%. We assume that the ionic liquid has penetrated the nanosheet film and a quasi-static state is reached, which is corroborated by the relatively stable and repeatable transfer curves (Figure S7, Supporting Information).

Cyclic voltammetry (CV) was used to investigate charge transfer to and from the network and enabled the determination of \( C_v \). For these measurements, the source and drain contacts were shorted externally to form the working electrode and the displaced (side-) gate electrode acted as the counter electrode. An oxidized silver wire was immersed into the IL above the channel and used as a pseudo-reference electrode.[50] Using this configuration, the potential drop across the working electrode, \( \Delta V_{s-ch-d} \), and the resulting current, \( I_g \), were monitored.

The CV traces shown in Figure 3c correspond to different voltage sweep rates (\( d\Delta V_{s-ch-d}/dt = 20–100 \text{ mV s}^{-1} \)) for the same device as for Figure 2. We have presented the CV data as \( -I_g \) versus \( -\Delta V_{s-ch-d} \) (unlike standard electrochemical conventions), such that hole and electron accumulation regions mirror those of standard transistor measurements (i.e., hole and electron accumulation). The potential limits were chosen to approach the ideal polarizable window of the electrolyte.

The CVs feature a window of relatively constant current flow corresponding to the bandgap region of the WS\(_2\). Here, the charge accepted is mainly due to the interfacial capacitance of the source and drain contacts as well as possible mid-gap states. Figure S13a (Supporting Information) shows the CV response of only the source and drain contacts in comparison to the same structure with an overlying WS\(_2\) network. For potential excursion beyond \( -\Delta V_{s-ch-d} = 0.4 \text{ V} \), we observe significantly increased current indicating filling of a greater density of electronic states, approaching the conduction band of the network. While we cannot designate the precise origin of these potential dependent features, we note that they are absent from the CV measurements for the IL-gated single-crystal multilayer WS\(_2\) channel prepared by Braga et al.[34] These show more ideal looking, trap-free characteristics with a pair of sharp peaks separated by an energy close to theoretical bandgap of WS\(_2\) (\( E_{g} = 1.3 \text{ eV} \)).[52] Indicating the filling (depletion) of the conduction (valence) band states. Hence, the additional features present for our network-based EGTs imply the presences of a range of additional electronic (trap) states. These are likely to be responsible for the large subthreshold slopes and onset voltages observed for transfer measurement. Repeated CVs were performed (350 cycles) to verify that the current response is stable (Figure S13b, Supporting Information).

The sweep rate dependence of the CVs (Figure S14, Supporting Information) shows that while \( I_g(\Delta V_{s-ch-d}) \) varies linearly across the potential window, as required for capacitive processes, the capacitance is strongly potential dependent. Thus, to arrive at an estimate for \( C_v \), we integrated \( I_g \) over a wide potential range to yield an average volumetric capacitance under electron accumulation, as shown in Equation (5).

\[
I_g = \frac{\mu_{sc} \cdot t_{ch} \cdot W \cdot C_v}{2L}(V_g - V_{th})^2 \quad (4)
\]

These equations differ from their standard forms (for field-effect transistor within the gradual channel approximation) due to the introduction of the volumetric capacitance, \( C_v \), and channel thickness, \( t_{ch} \). This modification is needed, as we expect that charge accumulation occurs throughout the volume of the porous channel material when electrolyte-gated.[69] rather than solely at the surface of the network. The transconductances were determined directly (Figures S10 and S11, Supporting Information) and \( C_v = C/(A t_{ch}) \) is the volumetric capacitance of the nanosheet network (as discussed below). The film thickness was measured for each sample by contact profilometry; the devices exhibited a mean thickness of \( t_{ch} = 150 \pm 20 \text{ nm} \) (Figure S8, Supporting Information). The footprint area, \( A \), of each device was measured by optical microscopy (Figure S12, Supporting Information). The uncertainty in the thickness and precise area of the porous film increases the error for the estimated carrier mobility, although by less than 20%. We assume that the ionic liquid has penetrated the nanosheet film and a quasi-static state is reached, which is corroborated by the relatively stable and repeatable transfer curves (Figure S7, Supporting Information).

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The lower integration limit was set to the middle of the bandgap region (−V_{s-ch-d} = 0.35 V), and the upper limit set prior to the onset of Faradaic current flow (these give a voltage window of ΔV = 1.85 V). Using this approach, we find C_V close to 30 F cm⁻³ for all samples (Figure 3d) decreasing only slightly as a function of sweep rate (Figure 3c, inset). This approach yields the maximum capacitance that can be expected from these networks and enables calculation of a minimum charge carrier mobility using Equations (1) and (2). As shown in Figure 3e, the obtained mobility values are fairly modest (μ_{lin} = 0.013 cm² V⁻¹ s⁻¹ and μ_{tot} = 0.009 cm² V⁻¹ s⁻¹) and did not vary significantly among the sample set. Although few groups studying electrolyte-gating of porous thin film EGTs report volumetric capacitance, Rivnay et al. measured a similar value of C_V = 39 F cm⁻³ for PEDOT:PSS-based devices. While Kelly et al. reported a significantly lower value of C_V = 1.2 F cm⁻³, in this case the integration was confined to a narrow region of the potential window (ΔV = 0.2 V). For comparison of different EGTs, a reliable determination of C_V is necessary. Inal et al. also reported the product of mobility and C_V as a possible figure of merit to compare different EGTs, with values from 0.1 to 200 F cm⁻¹ V⁻¹ s⁻¹ for various conjugated polymers. In comparison, the presented WS₂ nanosheet networks reach only about 0.5 F cm⁻¹ V⁻¹ s⁻¹. Such low values reflect the relatively poor mobilities of these networks that rely on charge hopping between many flakes.

For all transistors the relative contributions of contact and channel resistance are important. Hence, output measurements were performed as a function of channel length to determine the contact resistance. Figure 4a presents the output curves for a typical sample at low bias conditions (V_d < 0.1 V) and for a range of gate voltages (V_g = +1.6 to +2.0 V). The total resistance was extracted from the slope, R_{tot} = dV_d/dI_d and plotted as a function of channel length in Figure 4b and gate voltage (see inset). Unexpectedly, we observe a superlinear increase with channel length, which may suggest a lateral electric field (E) dependence of charge transfer between the nanosheets. Similar behavior has been observed for a range of disordered semiconductor systems and is often interpreted in terms of a Poole–Frenkel process (μ ∝ exp(E_{1/2})). This behavior will be investigated more closely later. Although the “transmission line method” is strictly not applicable under these circumstances, the channel length data is sufficiently linear for L ≤ 10 µm to allow for a rough extrapolation to L = 0 and an estimation of R_c with 270–800 Ω cm depending on gate voltage (see Figure 4c). The obtained values are similar to those reported for other EGTs.

To investigate the impact of the lateral electric field on charge transport further, the electron density in the channel should be kept constant (as in the linear regime), while increasing the drain voltage and thus the lateral electric field. At room temperature, increasing the drain voltage (and thus change of the potential difference to the gate) leads to a redistribution of ions and thus also of the accumulated electrons in the channel.
transport in layered semiconductor networks processed in solvents (such as isopropanol) or uncharged aqueous stabilizer systems, which might indicate whether the anionic surfactant retained in these films plays an important role for this hopping mechanism.

The temperature-dependent on-conductance for a range of $V_d$ (Figure 5c) was further fitted to an Arrhenius-type relation, $G \propto \exp(-E_A/kT)$ to yield an apparent activation energy $E_A$ for both channel lengths (Figure 5d). Note that the extracted activation energy should really be the sum of $-E_A$ and $\gamma E_{\text{F}}^{1/2}$ as shown in Equation (6) and thus depend on the electric field. Indeed, we observe a slight decrease of $E_A$ with drain bias over the tested voltage range and a substantially larger value for the long channel EGT (lower lateral electric field) under otherwise equivalent conditions. These observations are clear evidence that charge transport within these nanosheet networks depends on the lateral electric field in addition to thermally activated hopping. Further experiments are required to determine the precise mechanism. Interestingly, the extracted apparent activation energies are relatively low (less than 50 meV in all cases), and similar to that obtained for transport within solution-processed graphene networks embedded in a polystyrene binder ($E_A = 56 \text{ meV}$). An even lower value ($E_A = 15 \text{ meV}$) was found for rGO nanosheets with h-BN insulating barriers. Here, it was suggested that oxide groups at the BN flake edge provide a reduced hopping barrier between adjacent rGO sheets. In all cases, such low activation energies may suggest that the large number of junctions is the main limiting factor for charge transport. Alternatively, the transport in these networks—similar to graphene networks—might be best described by the fluctuation induced tunnelling model, which leads to a rather flat temperature dependence at low temperatures.

Given that field-effect transistors based on individual WS$_2$ monolayered flakes show fairly high mobilities (tens to up to hundreds of cm$^2$ V$^{-1}$ s$^{-1}$), the most significant factor leading to the unimpressive overall mobilities of these nanosheet networks should clearly be the need for hopping between charge carriers between flakes. Therefore, lateral dimensions of the nanosheets should be maximized to improve performance, as was shown for rGO networks. While we have made efforts to maximize the mean lateral flake size (within the limitations of standard liquid phase exfoliation), the nanosheets are also quite thick (due to $<N> \propto <L>$). Very little is known about the influence of flake size and thickness on the transport properties of nanosheet networks, let alone the potential influence of morphology when produced by different deposition methods, which may influence interflake contact, flake overlap area etc.

As such, we have also explored the performance of EGTs prepared from nanosheet networks with other mean flake geometries. To do so, two other WS$_2$ nanosheet dispersions were prepared and the average flake size was varied using a typical centrifugation cascade. The resulting mean nanosheet sizes were determined using UV-vis extinction spectroscopic metrics (see Figure S1 in the Supporting Information). These dispersions featured mean length and mean layer number combinations of $<L> = 220 \text{ nm}$, $<N> = 14$ and $<L> = 110 \text{ nm}$, $<N> = 8$, respectively. EGTs with various channel lengths were prepared from these dispersions in the same manner as for the larger flake dispersion. Figure 6a,b presents transfer measurements as shown in Figure 5a for the $L = 35 \mu$m device and in Figure S15 (Supporting Information) for the $L = 2 \mu$m device.

We observe a superlinear increase in current with drain bias, i.e., increasing conductance as a function of the applied lateral electric field across the channel as well as thermally activated transport (drain current increasing with temperature). The increase of current with lateral electric field again points toward a field-assisted transport, such as in the Poole–Frenkel model. For disordered semiconductors the temperature and field dependent electrical conductivity, $\sigma$, can often be described by a version of Equation (6)

$$\sigma = \sigma_0 \cdot \exp \left[ \frac{\gamma E^{1/2}}{kT} \right] \cdot \exp \left[ -\frac{E_A}{kT} \right]$$

Here, $\sigma_0$ is the conductivity in the absence of an electric field, $k$ is the Boltzmann constant, and $\gamma$ is a material-dependent factor. The Poole–Frenkel mechanism involves the lowering of the potential barrier that a charge carrier has to overcome to leave a charged trap by the applied lateral field. This might be a reasonable assumption due to the presence of residual cholate in the nanosheet film. By re-expressing the data in Figure 5a as $\ln(I_d/V_d)$ versus $V_d^{1/2}$ (Figure 5b), the observed linearity indeed indicates that a Poole–Frenkel-like hopping mechanism is a possible explanation for the observed transport behavior in these networks (at least within this temperature range). Similar behavior was reported for reduced graphene oxide (rGO) networks interspersed with few layer insulating (h-BN) nanosheets. Future experiments should explore...
indicating that nanosheet networks based on medium flake size \( <L> = 220 \text{ nm} \)—although inferior—are not substantially different from larger flake networks despite the mean lateral size of the flake being close to half. On the other hand, transport within the smallest flake networks is rather impaired, with lower on-currents and increased threshold voltages. Interestingly, p-type transport becomes more pronounced for smaller (and thinner) flakes. The reasons for this effect are unclear. As expected the width-normalized on-conductances \( G_{on}/W \) are significantly reduced for smaller nanosheet sizes (see Figure 6c,d) and the device-to-device variability increased. These results suggest that there is still much to be learned about charge transport within solution-processed semiconducting nanosheet networks in order to improve effective mobilities and on-conductances.

3. Conclusion

We have demonstrated the first n-type electrolyte-gated transistors based on solution-processed WS\(_2\) nanosheet networks. The layered WS\(_2\) inks were produced by liquid phase exfoliation in aqueous/surfactant media and deposited at low temperatures \( (T = 120 \degree C) \) by inexpensive and scalable airbrush deposition, without the need for solvent exchange, further additives or complicated post-processing. This simple, scalable and low-cost process, however, came at the expense of device performance compared to single flake WS\(_2\) transistors. The network mobilities were fairly low \( (\mu_{\text{lin}} \leq 0.013 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}) \), although they exhibited reasonable width normalized on-conductances of \( G_{on}/W = 0.27 \mu \text{S} \mu \text{m}^{-1} \) and on/off ratios of \( 10^4 \). Some p-type transport was also observed but varied considerably among devices. The volumetric capacitance of these networks was fairly large \( (C_V = 30 \text{ F cm}^{-3}) \), and CV measurements indicated a significant proportion of trap states. Charge transport in the disordered nanosheet films was clearly dependent on the applied lateral electric fields and thermally activated. A cursory investigation of flake size dependence confirms the need for flakes with large lateral dimensions to improve conductivity. Future studies should focus on improving interflake charge transfer and the reduction of trap states to reach sufficient device performance for practical applications while maintaining low cost and processability.

4. Experimental Section

Dispersion Preparation: WS\(_2\) powder (Sigma Aldrich, Lot STBF7189V) was added to an aqueous solution of sodium cholate (SC, Sigma Aldrich, Lot SLBQ4966V): the initial concentrations were \( C_{WS2} = 30 \text{ g L}^{-1} \) and \( C_{SC} = 6 \text{ g L}^{-1} \) and total volume was \( V = 80 \text{ mL} \). The mixture was probe sonicated using a flathead tip with replaceable end (13 mm diameter) for 1 h at 60% amplitude with a 4 s/4 s on/off pulse (Sonics, Vibracell VXC-500) in a metal beaker. The temperature surrounding the sample beaker at \( 5 \degree C \) was maintained at \( 5 \degree C \) by a chiller system. The mixture was centrifuged for 1.5 h at 5000 g using a Beckman Coulter Avanti J-26XP centrifuge equipped with a JA-25.50 rotor. The supernatant was discarded and the sediments combined. Fresh dispersant (at reduced SC concentration, \( C_{SC} = 2 \text{ g L}^{-1}, V = 80 \text{ mL} \)) was added to the combined sediment and the mixture was sonicated for 7 h (as before, but 6 s/8 s on/off pulse). Centrifugation based size selection was then performed to obtain three separate dispersions with different average flake characteristics\([19]\). The trapping window for each dispersion was 100–400 g, 400–800 g, and 800–2000 g. Here, the supernatant from a given step (e.g., after 100 g) was used for the following centrifugation (i.e., 400 g) and sediment obtained became a new dispersion. The supernatant was then centrifuged at higher rotation (i.e., 800 g). The centrifugation time in each case was 2 h. The sediments were redispersed to 0.4 g L\(^{-1}\) using DI water by bath sonication for 2 min (Branson 2510 bath sonicator).

Dispersion Characterisation: UV–vis extinction spectra were measured using a 1 cm quartz cuvette from 200 to 800 nm (data resolution set to 0.5 nm) using a Cary 6000i spectrometer (Varian Inc.).

Nanosheet Network Film Preparation and Characterization: Glass substrates (Schott AF32 Eco, 25 \( \times \) 20 mm\(^2\)) were cleaned by bath sonication in acetone and isopropanol for 10 min each, rinsed with DI water, and dried using compressed nitrogen. The cleaned substrates were patterned with interdigitated channel contacts (source and drain) and a pair of displaced gate electrodes using standard double-layer resist photolithography, e-beam evaporation (Cr/Au with 2/30 nm thicknesses) and lift-off. The structure makes available multiple devices with variable channel lengths \( (L = 2–35 \mu \text{m}) \) and a constant channel width of \( W = 1 \text{ cm} \). The two gate electrodes provide a large combined gate electrode with area of \( A = 0.94 \text{ cm}^2 \). The patterned substrates were cleaned by bath sonication in acetone and then isopropanol at \( 60 \degree C \) for 10 min each. Finally, the substrates were rinsed with DI water and then dried on a hot plate at \( 160 \degree C \) for 5 min.

Porous nanomaterial networks of layered WS\(_2\) were deposited onto the interdigitated source-drain contacts by airbrush spraying of the nanosheet dispersions. The dispersions \( (C_{WS2} = 0.4 \text{ mg mL}^{-1}) \) were bath sonicated for 2 min prior to deposition using a Branson 2510 sonicator. Film deposition was achieved using an Infinity CR Plus airbrush (Harder & Steenback) fitted with a 0.15 mm nozzle. Compressed N\(_2\) was used to atomize the dispersion (1.0 bar) and the needle displacement upon triggering (controlling the orifice area) was set to 150 \( \mu \text{m} \). The substrate
was placed on a heated stage (T = 120 °C) located 10 cm from the nozzle, and the stage was rastered back and forth through the spray at 3 cm s⁻¹. A laser-patterned stainless stencil (Becktron) was placed over the substrate to confine the deposited material to a stripe, which was further patterned by hand using a toothpick. The film thickness was determined by the volume of dispersion sprayed (at CWS = 0.4 g L⁻¹). After deposition, the substrate was soaked in DI water for 12 h and then dried in vacuum (pressure = 800 mbar). Samples were then moved to a N₂ glove box and heated on a hotplate at T = 100 °C for 12 h. The nanosheet film morphology was imaged using a JEOL JSM-7610F field emission scanning electron microscope (FE-SEM). Film thickness profiles were obtained using a Bruker DektakXT stylus profiler.

**Device Characterization:** All electrical and electrochemical measurements were performed either inside a dry nitrogen glove box or under vacuum (<10⁻⁶ mbar) in a CRX-6.5K probe station (Lake Shore Cryotronics). In the former case, a Dow Corning PDMS frame was prepared using Sylgard 184 base/curing agent kit and attached to the substrate to contain the ionic liquid electrolyte (Merck KGaA supplied 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide, [EMIM] [TFSI]). Current voltage measurements were performed using an Agilent 4155C semiconductor parameter analyser. Cyclic voltammetry was performed using an Autolab PGSTAT204 potentiostat whereby the combined gate electrodes were used as the counter electrode and source–channel–drain region as the working electrode. The source and drain electrodes were contacted individually and connected by cable. For these measurements, an oxidized silver wire pseudo-reference electrode was suspended in the ionic liquid above the working electrode.

**Supporting Information**
Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**
The authors declare no conflict of interest.

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electrolyte-gating, field-effect transistor, semiconducting nanosheet network, transition-metal dichalcogenide, tungsten disulfide

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