Electronic properties and circuit applications of networks of electrochemically exfoliated 2D nanosheets

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Abstract:

High aspect-ratio 2D materials are promising for solution-processed electronics, yet the factors controlling exfoliation remain unclear and relatively few solution-processed networks have been electrically characterized. Here we combine theory and experiment to show that electrochemical exfoliation of layered crystals with sufficient stiffness-anisotropy (in-plane/out-of-plane Young's modulus ratio >1.7) yields high aspect-ratio nanosheets with intrinsic mobilities μ_{NS}=20–75 $cm^2V^{-1}s^{-1}$ across transition metal dichalcogenides and related alloys. Impedance spectroscopy indicates that solution-deposited networks can achieve junction-to-nanosheet resistance ratios (R_J/R_{NS}) as low as ~3, supporting theoretical predictions that $\mu_{NS}/\mu_{Net}=R_J/R_{NS}+1$ and suggesting that further reductions in R_J will increase μ_{Net} toward the nanosheet limit (μ_{NS}). These networks display n-type, p-type, and ambipolar behaviour, with on/off ratios up to 10^5 and mobilities μ_{Net} = 13 $cm^2V^{-1}s^{-1}$. Here, we show that such high-performing 2D materials enable functional solution-processed circuits, including inverters, buffers, a 4-bit digital-to-analog converter, and a circuit capable of encoding and decoding 7-bit ASCII messages.

Introduction:

Solution-processed electronics are important for circuits requiring low-cost components and manufacturing scalability (m^2/minute) on a wide range of non-conformal substrates (e.g. textile and polymers).[1,2] Two-dimensional (2D) semiconducting nanosheets show great promise in this area due to their solution-processability,[3,4] diversity[5], mechanical flexibility[6], and impressive intrinsic electrical properties (e.g. nanosheet mobility, μ_{NS}>50 $cm^2\ V^{-1}\ s^{-1}$)[7]. Liquid-phase exfoliation[8,9] (LPE) has yielded many types of semiconducting nanosheets with low aspect ratio (AR < 30).[8] These have been solution-processed into electronic devices with poor performance, displaying network mobilities, $\mu_{Net} \sim 0.1\ cm^2\ V^{-1}s^{-1}$ and current on/off ratio, $I_{on}/I_{off} \sim 10^2$, significantly below expectations for individual nanosheets.[7,10] Conversely, electrochemical exfoliation[4,11,12] (EE) is a method known to produce high AR nanosheets,[13-15] leading to high mobility networks (μ_{Net} up to 15 $cm^2\ V^{-1}s^{-1}$),[4,16-18] due to the link between high AR and low junction resistance (R_J).[19] In addition, transition metal phosphorus trichalcogenides (M-P-X_3, TMTs)[20], transition metal monochalcogenides (M-X, TMMs)[13] and elemental materials[21] have been electrochemically exfoliated although the electrical performance of their networks are unknown. Moreover, the factors determining successful exfoliation remain unclear in general.

The AR of nanosheets produced by LPE depends on the balance between in-plane tearing energy and out-of-plane peeling energy.[8] In EE, it is expected that ion insertion lowers the peeling energy, aiding exfoliation and maximising AR. Increased AR has been linked to the formation of conformal, low-resistance junctions within networks.[19] However, the influence of the crystal's mechanical properties on the length (L) and thickness (t) of electrochemically exfoliated nanosheets is unstudied. We believe a deeper understanding of the physics of the exfoliation process could facilitate nanosheet optimisation, increase aspect ratios and so minimise R_J.[19,22]

For networks, theory predicts[22] that $\mu_{NS} / \mu_{Net} \approx R_J / R_{NS} + 1$. Thus, reducing R_J below the resistance of the individual nanosheets (R_{NS}) would result in μ_{Net} approaching its upper limit of μ_{NS},[22] opening the way toward high-performance solution processed devices. However, for most electrochemically exfoliated nanosheet types, the actual values of μ_{NS} are unknown as are values of R_J in solution-processed nanosheet networks. This means that for most types of 2D materials, we have no idea what the performance limits are or how far we are from achieving them.

Furthermore, although significant progress has been made recently in solution-processed logic with complementary functionality[23-26] and resistive random access memory,[27,28] we still lack important solution-processed circuits, such as digital-to-analogue converters (DACs), commonly used in microcontrollers and systems-on-chip to convert digital signals into continuous analog signals, and binary-amplitude-shift-keying (BASK) circuits, used in digital communication and signal processing.[29] The development of solution-processed, 2D-based DACs and BASK circuits would enable the next significant step in solution processed circuits.

Using known exfoliation protocols, we combine theory and experiment to explore the underlying mechanisms of crystal expansion in a range of materials. For each exfoliated material, we solution-deposit nanosheet networks, performing electrical, optical and transistor characterisation. For a subset of materials, we examine the factors limiting charge transport in these networks by measuring R_J, R_{NS} and μ_{NS} and using a network model to link these values to μ_{Net}. Finally, we use our best performing transistors to fabricate solution-processed resistor ladder networks, inverters and buffer circuits.

Results:

Selection of two dimensional material families: We initially choose twenty-eight low optical bandgap (E_{Op} < 2eV) 2D materials, a group which will be subjected to a rigorous down-selection process to identify champion materials for solution-processed circuits (Figure 1a). Figure 1b shows optical images of the 2D crystals grown by us and which can be grouped into five families: TMMs, TMDs, TMTs, a layered bismuth oxychalcogenides (BCT) and elemental materials (metalloids).

The TMMs are outlined in blue and are combinations of a chalcogen (S, Se or Te) with a group 13 (In, Ga, Tl) or group 14 (Sn, Ge) element. These elements are post-transition metals except for Ge, which is a metalloid. The TMDs, outlined in orange, combine one transition metal (W, Mo, Pt, Pd, Ta) or post-transition metal (Sn) and two chalcogen atoms (S, Se). We will also examine the alloy, $Mo_{0.5}W_{0.5}Se_2$, as well as substitutional doping of $MoSe_2$ and WSe_2 with niobium ($MoSe_2$:Nb, WSe_2:Nb) to induce p-type doping.[30] TMTs (green outline) such as $CoPS_3$, $FePS_3$, $NiPS_3$ and $HfSe_3$ will also be examined. We also examine elemental 2D materials of tellurene (Te), germanene (Ge), and black phosphorus (BP), which are shown with a red outline. Finally, BCTs such as bismuth oxyselenide Bi_2O_2Se (yellow outline) have shown great promise as high electron mobility materials (>100 $cm^2\ V^{-1}\ s^{-1}$) and is also included.[31]

Mechanical criteria for electrochemical exfoliation: To identify the factors controlling the electrochemical exfoliability of 2D materials, we use density functional theory to calculate the in-plane (E_{in}) and out-of-plane (E_{out}) Young's moduli as well as the inter-sheet binding energy, E_b, as described in Supplementary Note 2. We use, the ratio of E_{in}/E_{out} as a figure of merit for determining crystal expansion. In Figure 1c, we plot the E_{in} and E_{out} for each material where the dashed line represents where $E_{in}/E_{out} = 1$. Alternatively, it was previously proposed that layered crystals with E_b<120 meV Å^{-2} are potentially exploitable.[32,33] We use the materials cloud two-dimensional crystals database to determine the E_b of each material (Supplementary Note 2). We find that every crystal in this study has E_b < 90 meV Å^{-2}, suggesting that all of our 2D crystals are potentially exfoliable.

To assess the potential of E_{in}/E_{out} and E_b as metrics for electrochemical exfoliability, we have performed electrochemical exfoliation on each material.[11,34] We apply an electrical potential (8 V) to each crystal immersed in the electrolyte tetrapropylammonium bromide (TPA^+Br^-) (See Methods). Ideally, the intercalation of the TPA^+ cations swells the crystal, weakening the interlayer bonding, facilitating exfoliation by mild sonication. We emphasise that crystal expansion is a required prerequisite for exfoliation. In Figure 1d, we plot the calculated values of E_{in}/E_{out} and E_b as a phase diagram with the presence or absence of swelling indicated (green zone for swelling). We found expansion to occur for all crystals with $E_{in}/E_{out} > 1.7$, while for crystals with $E_{in}/E_{out} < 1$ limited expansion was observed, leading to very poor exfoliation (<0.01 mg nanosheets produced). However, while expansion could be correlated with $E_b < 40$ meV Å^{-2}, this criterion was not enough as crystals with E_{in}/E_{out} <1.7 showed limited or no expansion, indicating that $E_{in}/E_{out} > 1.7$ is a more basic requirement. Crystal interlayer spacing is uncorrelated with exfoliation efficiency (Supplementary Note 15). The expanded crystals were then liquid-exfoliated as described in the methods and then formulated into isopropyl alcohol (IPA) based inks as shown in Figure 1e. Optical absorption spectra for each ink are obtained using an integrating sphere and display the expected excitonic transitions further described in detail in Supplementary Note 8-12.[35]

High aspect ratio nanosheets are critical for solution processed electronics.[19] Atomic force microscopy (AFM) images such as those in Figure 2a allow the measurements of *L*, *t* and *AR* for individual nanosheets. Examples of *L*-*t* and *AR*-*t* data clouds are given for one material from each family in Figure 2b-c. Unlike LPE nanosheets,[8] there appears to be no *L*-*t* correlation within each material (Supplementary Note 14). The mean nanosheet length and apparent thickness are plotted for each material in Figure 2d (Supplementary Note 3). Most materials

have $\langle L \rangle > 1\ \mu m$ and $\langle t \rangle < 20\ nm$, although some materials are smaller and thicker (L < 1 μm, t < 100 nm). To probe the relationship between nanosheet dimensions and crystal properties, we plot $\langle AR \rangle$ versus E_{in} / E_{out} in Figure 2e. This shows clear evidence that crystals with stiffness anisotropy above a critical value, $E_{in} / E_{out} > 1.7$, yield nanosheets with AR > 100, underlining the importance of crystal mechanical properties.

High aspect ratios are required for nanosheets to form aligned films with conformal junctions as explained by Kelly et al.[19] Figure 2f shows examples of scanning electron microscopy images of solution-deposited nanosheet networks of materials that have not previously been electrochemically exfoliated for transistor device applications (Supplementary Note 14). High-AR materials such as InSe, $Mo_{0.5}W_{0.5}Se_2$ and $HfSe_3$ all have networks of well-aligned nanosheets with highly-connected junctions, requirements for low R_J.[19] In contrast, Te is a low-AR material and forms a much more disordered, particle-like network. Example Raman spectra are shown in figure 2g and are consistent with the expected semiconducting material phase and dimensions as described in detail in the Supplementary Note 4-7, 14, alongside information on the full characterisation of each material.

Network conductivity and transistor performance: Although nanosheet inks can be formed into networks in various ways, Langmuir–Schaefer (LS) coating is an efficient way to produce high-quality nanosheet networks. This process leverages the interfacial tension at a hexane/water interface to form highly aligned nanosheet networks with minimal (~120 μL) use of ink, as detailed in the Methods. We use this technique to fabricate networks (thickness t_{NET} ~ 19 - 50 nm, Supplementary Note 17) of 22 different nanosheets types on flexible polyethylene terephthalate (PET) substrates for electrical characterisation. Depending on the material, we either evaporated gold top electrodes or used pre-patterned electrode arrays (See Methods). In all cases, networks are annealed at 120 °C for 1 h in an inert N_2 environment.

Figure 3b plots σ_{Net} for each material and we estimate E_{Op} via Tauc plots with absorption spectra (Supplementary Note 13). With the exception of the Nb-doped TMDs and MoS_2 and $SnSe_2$, materials with higher E_{Op} tended to have lower σ_{Net}, indicating the importance of thermal carrier generation.[36] For example, despite the quality of their junctions (figure 2f), $HfSe_3$ and InSe have low conductivity due to their high bandgap. Otherwise, the spread in data implies a range of network mobilities from material to material. Broadly, TMDs are the most conductive (σ_{Net} >10^{-3} S/m) and TMTs the least conductive (σ_{Net} < 10^{-4} S/m), possibly due to

TMTs possessing anisotropic nanosheet electrical properties.[37] Similarly, the elemental nanosheet networks of BP and tellurene are anisotropic, resulting in $\sigma_{Net} < 10^{-3}$ S/m.[38,39] In materials with anisotropic electrical properties, charge carriers must traverse directions with lower conductivity, which increases the effective resistance of the network and lowers σ_{Net}.

Many applications in flexible electronics will require strain-independent electrical properties. We demonstrate this for an LS-deposited TaS_2 network in Figure 3c, finding a minimal resistance change of only ~1% per % of strain, i.e $\Delta R / R_0 \approx \varepsilon$. Because the piezoresistive response of any material (resistivity, ρ, Poisson ratio ν) is given by[40] $\Delta R / R_0 = (\varepsilon / \rho)\mathrm{d}\rho / \mathrm{d}\varepsilon + (1+2\nu)\varepsilon$, this implies that these networks have near strain-independent resistivity ($\mathrm{d}\rho / \mathrm{d}\varepsilon \sim 0$) and ν≈0, properties that are near ideal for strain-invariant devices. We demonstrate this by applying a cyclic strain to the TaS_2 network (figure 3c, inset), finding only a very small ripple on the film resistance. It is likely that this behaviour is a feature of all EE nanosheet networks, provided that the networks have similar structure, dimensionality and inter-nanosheet junctions.[41]

To further characterise the electronic properties of our semiconducting networks, we fabricated electrochemical transistors, as shown schematically in Figure 3d. We used a gold side gate for the electrodes and 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide (EMIM TFSI) to switch the channel.[42] The electrical characterisation is performed using a probe station in ambient air under atmospheric conditions. We sweep a gate voltage, V_G, from 3V to -3V and measure the transfer characteristics for each network at a drain-source voltage of V_{DS} = 1V. Networks with $\sigma_{Net} < 10^{-4}$ S/m (shown by the dashed red line in Figure 3b) could not be gated since their σ_{Net} was lower than the ionic conductivity of EMIM TFSI, making it impossible to distinguish between semiconductor modulation or electrolyte effects. For all other materials, we observed ionic gate currents ($I_G \sim 10^{-5}$ A, 5.2×10^{-9} A/μm, matching expected values from the literature)[43] below the measured drain current (I_D), confirming the drain current modulation to be due to carrier concentration changes within the semiconducting channel (Supplementary Note 18).

We observe n-type behaviour for $PtSe_2$, WS_2, and MoS_2, which is consistent with previous reports (Figure 3e).[44,45] The networks of $MoSe_2$, WSe_2, $MoTe_2$ and $Mo_{0.5}W_{0.5}Se_2$ display ambipolar behaviour, in line with expectations (Figure 3f).[6,46] Moreover, we observe p-type

behaviour for $MoSe_2$:Nb, WSe_2:Nb, tellurene and BP. The addition of four p-type networks is particularly noteworthy as there are relatively few p-type 2D solution-processed networks in the wider literature (Supplementary Note 29). P-type behaviour was also observed for GaTe and InTe although at extremely low μ_{Net} (Supplementary Note 16).

Figure 3h shows a summary of the average μ_{Net} and I_{on}/I_{off} for our electrochemically gated transistors, with the best performing transistors found at the top right-hand corner of the plot. We obtain average μ_{Net} in the range of 1 - 8 $cm^2\ V^{-1}\ s^{-1}$ and average peak mobilities μ_{peak} >10 $cm^2\ V^{-1}\ s^{-1}$ for TMDs with I_{on}/I_{off} in the range of 10^0 - 10^5 (Supplementary Note 17). In some cases, relatively high off-currents were found as is often observed during electrochemical gating. The best performing family was the TMDs, where WS_2, WSe_2:Nb and $Mo_{0.5}W_{0.5}Se_2$ had complementary n-type, p-type and ambipolar properties, respectively. Substitutional doping proved a good strategy to convert the ambipolar materials, WSe_2 and $MoSe_2$, to p-type while retaining reasonable μ_{Net}.

In Supplementary Note 29, we have compiled a comprehensive list of state-of-the-art solution processed 2D semiconducting networks and their performance to date. We stress the realization of network-based transistors with materials that have been rarely applied for solution processed electronics: $WMoSe_2$, $MoTe_2$, tellurene, InTe, GaTe, $MoSe_2$:Nb and WSe_2:Nb, while demonstrating high μ_{Net} and I_{on}/I_{off} for some of them. Our devices show μ_{Net} and I_{on}/I_{off} comparable to some of the best organic polymers,[47] carbon nanotubes[48] and metal oxides[49] (μ_{Net} ~ 10 $cm^2\ V^{-1}\ s^{-1}$ with $I_{on}/I_{off} < 10^5$).[6] Furthermore, our μ_{Net} and I_{on}/I_{off} are higher than what has been achieved for LPE semiconducting transistors, typically μ_{Net} ~ 0.1 $cm^2\ V^{-1}\ s^{-1}$ with $I_{on}/I_{off} < 10^3$.[50,51] While our devices are comparable to the best EE networks μ_{Net} ~ 10 with $I_{on}/I_{off} < 10^6$, we demonstrate a much broader range of materials with complementary electrical properties. The majority of the literature has only examined MoS_2 networks for transistors, with a few exceptions beyond it.[18,25,52,53] We attribute the high μ_{Net} and I_{on}/I_{off} to our use of crystals with favourable mechanical properties ($E_{in}/E_{out} > 1.7$), which enables electrochemical exfoliation of high $AR > 10^2$ - 10^3 nanosheets leading to networks with low R_J.[22] Our devices represent a significant broadening in the library of complementary 2D nanosheets available for solution-processed circuits.

We can combine the network conductivity reported in Figure 3b with the mobility values in Figure 3h to obtain carrier densities ($\sigma_{Net} = nq\mu_{Net}$) as shown in Figure 3i. We find very high carrier densities in the Nb-doped materials, but also in MoS_2 and $PtSe_2$. In $PtSe_2$, this can be

attributed to the low bandgap. However, in MoS_2, this suggests a significant degree of inadvertent doping and explains its position in Figure 3b as well as its high I_{off}. The *n*-values found are as expected and we explain their positions relative to each other in Supplementary Note 20.

Junction resistance governs **network mobility:** Modelling[22] shows that μ_{Net} and R_J are linked in nanonetworks via $\mu_{NS} / \mu_{Net} \approx R_J / R_{NS} + 1$. This means that the network mobility can be increased by reducing R_J / R_{NS}. In addition, it shows that μ_{Net} can never exceed μ_{NS}, with this maximum value realizable by achieving $R_J << R_{NS}$.[22] Thus, knowledge of μ_{NS} is important to know the upper mobility limit while knowledge of R_J / R_{NS} is needed to know how far a network is from this limit. Here we use optical-pump terahertz-probe spectroscopy (OPTP) and time-resolved terahertz spectroscopy (TRTS) on a select number of promising 2D EE networks to determine the nanosheet mobility, μ_{NS} (Supplementary Note 21). The results of TRTS are shown in Fig. 4a, which range from 20 – 80 $cm^2\ V^{-1}\ s^{-1}$ and are compared to the measured network mobilities in Figure 4b. We find that in all cases, $\mu_{Net} < \mu_{NS}$, confirming the devices are junction limited. The tungsten-based samples, WSe_2 and WS_2 have higher μ_{NS} than the molybdenum-based samples, MoS_2 and $MoSe_2$, suggesting lower defect contents (Supplementary Note 21). The higher μ_{NS} for the selenide-based samples than sulphur-based samples could be due to the former being less defective. Nb-doping appears to reduce the mobility for both tungsten and molybdenum based samples, probably due to defect or impurity scattering.[54]

We have recently demonstrated that A.C. impedance spectroscopy allows R_J and R_{NS}, to be measured simultaneously,[22] although we only reported data for MoS_2. In this method, the measured network impedance is converted into the impedance of the average nanosheet-junction pair, $Z_{NS\text{-}J}$ (Supplementary Note 22). The real $Z_{NS\text{-}J}$ spectrum is then fitted with an equivalent circuit model to yield values of R_{NS}, R_J. Such spectra are shown in Figure 4c for networks of MoS_2, WS_2, $MoSe_2$ and $MoSe_2$:Nb with fits shown as solid lines (See Methods and Supplementary Note 22). The resultant R_J and R_{NS} data are shown in Figure 4d. The R_J values for our EE networks are >1000 times lower than reported values for LPE nanosheet networks (R_J = 6 - 24 GΩ),[22] emphasising the junction quality in EE networks. However, in all cases, we find $R_J/R_{NS} > 1$, indicating that the networks are junction-limited, and consistent with our conclusions from THz spectroscopy. As mentioned above, μ_{Net} and R_J are linked in nanonetworks via $R_J / R_{NS} \approx \mu_{NS} / \mu_{Net} - 1$.[22] As shown in Figure 4e, this relationship holds well

in our networks further emphasising the junction-limited nature of these systems. However, for MoS_2, we find R_J/R_{NS} =3.3, consistent with only mild junction limitations. In the future, junction resistances are likely to be further reduced by nanosheet optimisation and process engineering such that we anticipate that $R_J<R_{NS}$ is achievable, leading to $\mu_{Net} \sim \mu_{NS}$. To help understand the factors involved, we plot μ_{Net}/μ_{NS} versus mean nanosheet aspect ratio. With the exception of the MoS_2 and $Mo_{0.5}W_{0.5}Se_2$ data points, we see a well-defined trend where μ_{Net}/μ_{NS} increases super-linearly with aspect ratio in Figure 4f. This is probably a manifestation of the fact that increasing aspect ratio results in higher R_J (due to reduced nanosheet thickness) and lower R_{NS} (larger nanosheets should lead to larger junction area). Thus, increasing aspect ratio is a promising route to increased R_{NS}/R_J and so higher values of μ_{Net}/μ_{NS}.

Functional circuits from solution processed networks: Only a small minority of 2D materials produced by EE have been studied for circuit applications. Above, we surveyed a range of n, p and ambipolar 2D networks using electrochemically-gated transistor measurements because of the simplicity of this technique. However, due to their low ionic mobility, electrochemical transistors are often too slow (>100 ms response times)[10] to be used in practical circuits. Therefore, we select our most promising materials, WS_2 and $Mo_{0.5}W_{0.5}Se_2$, for use in solid state field-effect transistors (FETs) and circuits using native AlO_x as a dielectric (See Methods). We use these FETs to make a combination of digital-to-analog converters (DACs, Supplementary Note 26, 27), inverters (Supplementary Note 24) and buffer circuits (Supplementary Note 28) demonstrating circuits with complementary functionality.

The arrays of DAC circuits are shown in Figure 5a with optical microscopy (Leica DM6M microscope). The process involves using a binary weighted resistor network (*R*-2*R* ladder network) to convert a digital binary input into an analog output voltage.[55] Figure 5b magnifies the circuits and shows an optical image of the solution processed WS_2 4-bit ladder network and its corresponding circuit diagram. We use the solution-processed DAC to restore an analogue signal, as shown in the circuit diagram of Figure 5c. We input an analog triangular waveform, V_{IN}, into a conventional Si AD7819 analog-to-digital converter (ADC), which outputs a digital signal of logic "1" or "0". We first make a 3-bit DAC and connect the ADC to the DAC using bit lines (b_2, b_1 and b_0), where each bit represents a power of 2 in binary weighting. In our 3-bit DAC, b_2 represents the most significant bit and b_0 represents the least significant bit. We define V_{DD} as the power supply voltage of the ADC. If the value of a bit line is "1", the corresponding ADC output will go to the value of $V_{DD,}$ and if it is "0" the output will go to

ground. Figure 5d shows the digital signals of bits b_2 (red), b_1 (green), and b_0 (blue), and the corresponding binary weighted integer N. We achieve these weights by summating the contribution of each digital signal to the output voltage, V_{OUT}, of the DAC. The contribution of each signal is halved for each node going to the output. For example, in a 3-bit DAC, the contribution of b_2 is halved once, b_1 twice and b_0 three times. In Figure 5e, we show the V_{IN} (orange) to the ADC and the analog output of the WS_2 DAC (purple line). A more detailed explanation of the DAC operation can be found in Supplementary Note 26. We can also increase the complexity of the circuit by making a 4-bit WS_2 DAC that uses an additional binary weighted resistor (b_3) to give 16 discrete V_{OUT} values (Supplementary Note 27).

As an additional circuit demonstration, we use a digital signal, similar to the output of the ADC, and encode it into an analog signal using a binary amplitude shift keying (BASK) circuit, as shown in Figure 5f. We make a buffer using the manufacturing protocol of the NMOS depletion logic. In this case, the top $Mo_{0.5}W_{0.5}Se_2$ FET (driver) is used as a switch, and the bottom transistor is used as a depletion load. We provide a high-frequency (1 kHz) A.C. sine wave, with an amplitude of 200 mV, to act as a carrier signal denoted as the local oscillator, V_{LO}. The digital message is shown in the top of Figure 5g, as the V_{IN} to the gate of the top FET. When V_{IN} is -1 V, the top $Mo_{0.5}W_{0.5}Se_2$ FET is off and V_{OUT} is low and near zero volts. The top FET cannot be completely turned off, so there is a small amplitude signal (7.2 mV) at V_{OUT} according to voltage divider circuit analysis ($V_{OUT} = V_{LO} \times R_L/(R_{CH} + R_L)$), where R_{CH} and R_L are the channel resistances of the top and bottom FET, respectively. When V_{IN} = 0.87 V, the top $Mo_{0.5}W_{0.5}Se_2$ FET is on, and the carrier signal from the V_{LO} is passed to the output. Thus, we can encode the information from the digital signal into the amplitude of the high-frequency analog signal of $V_{LO.}$ We undertake a practical demonstration by demodulating a modulated encoded binary message from the high frequency signal using an AM demodulator (Supplementary Note 28). The message "Politecnico di Milano and University of Dublin" was encoded using 7-bit ASCII, resulting in 322 bits (data rate of 100 bits per second). Figure 5h shows the binary input to V_{IN} required to encode the message “Dublin”. The BASK circuit converts the binary sequence into an amplitude-modulated signal, shown as the orange curve in Figure 5h. The purple curve shows a 175 ms snippet of the decoded signal to reveal the letter “U” (Supplementary Note 28). These circuits help to understand the practical aspects of digital communication and signal processing in solution-processed 2D networks.

Discussion

We measure the electrical properties of low-bandgap 2D materials for solution-processed electronic applications. We determine that a high in-plane to out-of-plane stiffness ratio >1.7 and E_b < 40 meV Å^{-2} is essential to enable crystal expansion. Electrical characterisation of solution-deposited networks confirmed that a subset of these materials could achieve high mobility and I_{on}/I_{off} for both p-type and n-type 2D nanosheets. Measuring the nanosheet mobility for a range of materials (20 - 80 cm^2 V^{-1} s^{-1}), clarified the upper limit for network mobility. We decoupled the junction and nanosheet resistance for materials beyond MoS_2 and find the electrical performance is constrained by junction resistance (R_J/R_{NS}=3.3-23), which advances the understanding of performance limitations in 2D networks. Reducing these values below one will yield network mobilities approaching the intrinsic nanosheet limit. After a rigorous down selection of material electrical properties, we utilised WS_2 for the development of solution-processed DACs while $Mo_{0.5}W_{0.5}Se_2$ was utilised in a BASK circuit. A 7-bit ASCII message modulated with a solution-processed circuit and then decoded, which could be particularly useful for future solution-processed communication systems.

Methods:

Electrochemical Exfoliation: An electrochemical cell with two electrodes is utilized to intercalate the crystals. A small crystal piece measuring 0.1 × 1 × 1 mm serves as the cathode, while a platinum foil (Alfa Aesar) is employed as the anode. The electrodes are secured using copper crocodile clips. The electrolyte solution is prepared by dissolving tetrapropylammonium (TPA) bromide (5 mg/mL, Sigma-Aldrich) in approximately 50 mL of propylene carbonate. An 8 V voltage is applied across the electrodes for 30 minutes to intercalate the 2D crystal with TPA+ cations. Following this process, the 2D crystal expands to more than twice its original volume, confirming successful intercalation.

Ink Formulation: The expanded 2D crystal is subjected to bath sonication (Fisherbrand 112xx series) in a solution of 1 mg/mL poly(vinylpyrrolidone) (PVP, molecular weight ∼40,000) dissolved in dimethylformamide (DMF) for 5 minutes. This is followed by centrifugation (Hettich Mikro 220, 1195-A, radius 87 mm) at 500 rpm (24g) for 20 minutes to remove unexfoliated crystals. To size-select the dispersion, the supernatant (top 90%) is centrifuged at 1000 rpm (97g) for 1 hour, and the sediment is collected. To remove the PVP, the sediment obtained at 97g is diluted with 2 mL of DMF and centrifuged at 10,000 rpm (9744g) for 1 hour. This process is repeated twice, with the sediment being collected each time. A third washing step (to remove the DMF) involves diluting the sediment in 0.5 mL of isopropanol (IPA) and

centrifuging at 10,000 rpm (9744g), after which the sediment is collected. The final sediment is redispersed in approximately 0.5 mL of IPA, which is used in the study for each 2D crystal. **Raman Spectroscopy:** Inks of each material are drop cast onto Si/SiO_2 substrate and annealed at 120 °C. A Renishaw Raman spectrometer at 532 nm with a 100× objective is used to acquire spectra. An incident power of ~1 mW was used to minimise possible thermal damage. **Atomic Force Microscopy:** AFM measurements, including thickness and lateral nanosheet size analysis, are performed using a Bruker Multimode 8 microscope. The inks are diluted with IPA at a 1:100 ratio and then drop-cast onto silicon/silicon dioxide (Si/SiO_2) substrates. Post-dilution, the samples undergo an annealing process at 120°C for 30 minutes to evaporate any remaining solvent. For scanning, OLTESPA R3 cantilevers are employed in the ScanAsyst mode to systematically scan the samples. Approximately 25 to 50 nanosheets per sample are analyzed to gather statistical data. The lateral size of the nanosheets is determined by taking the square root of the product of the nanosheet's length and width. **Scanning Electron Microscopy**: SEM imaging was conducted using a Carl Zeiss Ultra SEM. A secondary electron detector was used to obtain the images at a 3 kV accelerating voltage, 5 mm working distance and 30 μm aperture.

Optical absorption spectroscopy: The spectra of nanosheet dispersions were collected by Cary 1050 spectrometer from 900 nm to 200 nm with a 1-2 nm step. The dispersion was placed in a 10 mm optical length cuvette. The absorption spectra were collected in an integrating sphere. The collected extinction and absorption spectra of nanosheet dispersions were subtracted by their corresponding solvent spectra to yield nanosheet only spectra. The scattering spectra were obtained by using extinction spectra and subtracting absorption spectra.

Langmuir–Schaefer Deposition: The Langmuir–Schaefer setup involves a Teflon stand where a PET substrate (2 × 2 cm, Novele, Novacentrix) (unless stated otherwise) is placed on top. The stand is placed in a 100mL beaker of deionized water. About 20 mL of distilled hexane is drop-cast onto the surface of the deionized water to create a water/hexane interface, under which the PET on the Teflon stand is submerged. Ink was drop-cast (~120 μL) onto the surface of the hexane until no gaps in the interface could be seen. A needle attached to a vacuum pump is used to extract the deionized water from the bottom of the Langmuir-Schaefer set-up, which extrudes the PET though the 2D crystal layer at a reproducible and consistent rate.[6] The process is repeated on separate PET substrates for every material to build the second layer of the network, except for MoS_2, which only uses one deposition layer. For GaTe, InTe, SnSe, and SnS networks, Si/SiO_2 substrates (Fraunhofer Gen 5 OFET chips) with pre-patterned gold

electrodes were used. The 2D nanosheet networks are annealed at 120 °C for 1 h on a hot plate in an N_2 glovebox (Jacomex GP campus) for each layer of deposition to remove residual solvent and improve the adhesion of the 2D nanosheets to the substrate.

Evaporation: Gold electrodes (~100 nm thick) are deposited by evaporation (FC-2000 Temescal Evaporator) through a stainless steel mask (50 μm thick), which is laser cut (Laser Micromachining ltd). The gold defines the channel dimensions of W_{CH} = 11000 μm and L_{CH} = 50 μm. The Au evaporation also defines our gate electrode, which is placed ~1 mm from the source and drain electrodes and is ~1.5 × 4 mm in size. These electrodes are used for the electrical characterisation presented in Figure 3 for all materials except GaTe, InTe, SnSe and SnS, which used pre-patterned gold electrodes from Fraunhofer. The Fraunhofer chips consisted of an array of 4 transistors with L_{CH} = 2.5 μm, W_{CH} = 10 mm and 230 nm SiO_2 thickness, with a gate of n-doped silicon ($n \sim 10^{17}$ cm^{-3}). We used the Fraunhofer chips for the electrical characterisation of these materials since the nanosheets were small < 500 nm, and therefore it was difficult to create a percolative network over a large L_{CH} = 50 μm made by the shadow mask. L_{CH} < 50 μm was not possible to achieve with the shadow masks without shorting the source and drain electrodes.

Stretchable Conductor: TaS_2 devices were tested by mounting samples in a Zwick Z0.5 Proline tensile tester with a 100 N load cell. The device was strained using a triangular sawtooth pattern at 2% strain, at a rate of 2 %/s. Conductive samples were contacted using conductive silver paint and silver wires. Two-probe electronic measurements were taken using a Keithley KE2601 source meter.

Impedance Spectroscopy: Fused silica substrates were purchased from MicroChemicals and used as the substrate for the impedance spectroscopy measurements. We use evaporated (FC-2000 Temescal Evaporator) Ti/Au (5 nm/95 nm) on the nanosheet network with a shadow mask with L_{CH} = 25 - 200 μm, W_{CH} = 19.4 mm. With a maximum frequency of 30 MHz, the Keysight E4990E analyser was used to obtain the impedance spectra. In order to minimise inductive artefacts at high frequencies, a test fixture (16047E) was utilised to connect the samples to the analyzer. This fixture allowed for a wire distance as short as 5 cm. Sensepeek SP10, a spring-loaded probe attachment, was utilised to link the analyzer to the substrates' contact pads. The spectra were obtained at a precision speed of three, with an amplitude of 500 mV. The network impedance, Z_{Net}, is converted into the impedance of the average nanosheet-junction pair, $Z_{NS\text{-}J}$ using the equation,[56]

$$Z_{NS-J} = \rho^{*}_{Net} \frac{(1-P_{Net})}{2t_{NS}} \left[1 + \frac{2}{n_{NS} t_{NS} l^{2}_{NS}} \right]^{-1} \quad (1)$$

ρ^{*}_{Net}, is the complex resistivity of the nanoheet network, $\rho^{*}_{Net} = Z_{Net}A/L_{CH}$, and A and L_{CH} are the network thickness and channel length. P_{Net} is the meso-porosity of the network previously found to be about once $P_{Net} \sim 0.02$, $< t >$, $< L >$ and n are known. The Z_{NS-J} spectrum is then fitted with an equivalent circuit model of a Randles circuit to yield values of R_{NS}, R_J.[56]

THz Spectroscopy: A titanium-doped regenerative amplifier (Libra), the foundation of our THz spectroscopy apparatus, generates 60 fs laser pulses with an 800 nm central wavelength. Optical photoexcitation of the sample, THz creation, and THz detection are the three separate uses for the amplifier's output. The first part of the beam is optically transformed by frequency doubling in a BBO crystal to a pump wavelength of 400 nm (photon energy 3.1 eV). Optical rectification at 800 nm is utilised in the second half to generate a THz waveform in a nonlinear ZnTe crystal with a duration of around 1 ps. The third component (at 800 nm) uses electro-optic sampling to detect the THz waveform after it has passed through the sample in a different ZnTe crystal. Mechanical delay stages regulate the time delays between the THz generation and detection pulse (t) as well as between the photoexcitation pump pulse and the THz detection pulse (τ). All of the experiments were carried out in a closed box with an N_2 environment at room temperature.

Transistor Measurements: We used a gold side gate (~1.5 mm × 4 mm) ~1 mm for the electrodes and 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide (EMIM TFSI) to switch the channel.[42] We used an FC-2000 Temescal Evaporator to evaporate Ti/Au (5 nm/95 nm) electrodes through a shadow mask with dimensions of L_{CH}= 50 μm and W_{CH} = 11 mm onto our Langmuir–Schaefer deposited networks. We control ion injection into our semiconducting channel using the ionic liquid 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide (EMIM-TFSI, Sigma-Aldrich). The ionic liquid is prepared by vacuum heating it at 100 °C for six hours in order to remove any absorbed water. The transistor is then carefully pipetted with a small amount of EMIM to make sure the gate and channel are sufficiently coated. The devices are vacuum-sealed for a full 12-hour period overnight in a Janis probe station in order to remove any residual water. The devices are then brought back to atmospheric pressure in order to be ready for measurements. We use a Keithley 2612A dual-channel source measurement instrument for electrical characterisation. Using a

scan rate of 50 mV s^{-1}, the transfer characteristics are carried out within a gate voltage window of -3 to 3 V. Additionally, throughout the measurements, V_{DS} is set to 1 V for every device.

Solid State electrostatic devices and electron-beam (e-beam) lithography: A fixed-beam moving-stage (FBMS) mode of a Raith eLINE system is operated at 30 keV to fabricate contacts for DACs, NMOS and BASK circuits. The FBMS mode allowed the patterning of very wide (650 μm) electrodes without stitching errors. Source and drain contacts for FETs used in NMOS and BASK circuits are 350 μm wide and are patterned in a 500 μm write field using a standard exposure mode. The electrodes were made of Ti/Au deposited in the e-beam evaporator at a base pressure of $\approx 1.2 \times 10^{-6}$ mbar. A thin native AlO_x layer, approximately 4 nm thick, formed on the surface of Al after the samples were exposed to air was used as the gate insulator.

Solution-Processed DACs: Highly doped Si/SiO_2 wafers (90 nm SiO_2) are utilised as substrates to fabricate the circuits. We extrude the wafers through a network of WS_2 nanosheets twice using the LS process. The WS_2 channels are defined by e-beam lithography and reactive ion etching (RIE) in SF_6 for 1 min at a power of 20 W. Next, nine electrodes were patterned to make an *R*-2*R* ladder network by e-beam lithography (W_{CH} = 200 and 300 μm for 2*R* and W_{CH} = 300, 400, 500 and 600 μm for *R*, with L_{CH} = 4 μm in all cases) followed by deposition of Ti/Au (3/80 nm) in an e-beam evaporator at a base pressure of $\approx 1.2 \times 10^{-6}$ mbar. The ADC used also has another input voltage (V_{REF}), which defines the maximum expected voltage at the analogue input (V_{IN_MAX}). For simplicity, we used $V_{REF} = V_{DD}$.

Solution-Processed NMOS and BASK circuit: First, we fabricate gate contacts by e-beam lithography and e-beam evaporation of Al (40 nm) onto the Si/SiO_2 wafers (90 nm SiO_2) and expose the samples to air to form a native AlO_x layer approximately 4 nm thick,[57] which serves as the gate oxide. The oxide capacitance per surface area is $C_{ox} \sim 1.4$ μF/cm^2, similar to previous work.[24] Next, we fabricate the source and drain contacts (L_{CH} = 1, 2, 3, 5 and 10 μm) of the FETs by e-beam lithography and e-beam evaporation of Ti/Au (3/37 nm). For the driver FET ($V_{GS} = V_{IN}$): W_{CH} = 300 μm and L_{CH} = 5 μm and for the load FET (V_{GS} = 0 V): W_{CH} = 300 μm, L_{CH} = 10 μm. We extrude the wafers through a network of $Mo_{0.5}W_{0.5}Se_2$ nanosheets twice using the LS process. The $Mo_{0.5}W_{0.5}Se_2$ channels (W_{CH} = 200 and 300 μm) are defined by e-beam lithography and RIE in SF_6 for 1 min at a power of 20 W.

Crystals: We prepare TMDs and TMTs by chemical vapour transport in quartz ampoules, sealed under high vacuum, with a series of controlled heating and cooling stages involving

two-zone furnaces to create thermal gradients for crystal growth. $PtSe_2$ and $PdSe_2$ are prepared by directly reacting elements in a stoichiometric ratio at high temperature (>800 °C) in a quartz ampoule. The BP is made by transporting red phosphorus in the presence of a mineraliser (SnI_4/Sn) in a quartz ampoule, sealed under a high vacuum and heated gradually in a muffle furnace. The TMMs are prepared by direct reaction of elements in a stoichiometric ratio in quartz ampoules, sealed under a high vacuum, heated above the melting point in a muffle furnace, and cooled slowly. The MoS_2 is of natural origin, and Bi_2O_2Se, thallium selenide (TlSe), tellurium, tin selinide (SnSe), tin sulfide (SnS) and germanium were bought commercially. Each crystal has a unique manufacturing protocol presented in further detail below. The crystals we will grow include, indium telluride (InTe), gallium telluride (GaTe), germanium sulphide (GeS), germanium selinide (GeSe), tungsten diselenide (WSe_2), molybdenum disulfide (MoS_2), molybdenum diselenide ($MoSe_2$), tungsten disulfide (WS_2), molybdenum ditelluride ($MoTe_2$), molybdenum tungsten diselenide ($Mo_{0.5}W_{0.5}Se_2$), platnium diselenide ($PtSe_2$), palladium diselenide ($PdSe_2$), tantalum disulfide (TaS_2), tin diselenide ($SnSe_2$), cobalt phosphorous trisulfide ($CoPS_3$), iron phosphorous trisulfide ($FePS_3$), nickel phosphorous trisulfide ($NiPS_3$) along with hafnium triselenide ($HfSe_3$).

Chemicals: Sulfur (99.9999%, 2-6mm), selenium (99.9999%, 2-4mm), tellurium (99.9999%, 2-6mm), phosphorus (99.9999%, 2-6mm), Ge (99.9999%, 1-6mm), Sn (1-4mm, 99.9999%), Ga (99.9999%, granules 1-6mm), In (99.9999%, 1-3mm) were obtained from Wuhan Xinrong New Materials Co., China. Tungsten (99.999%, -100 mesh) was obtained from China Rhenium Co., China. Niobium (99.9%, -100 mesh), tantalum (99.9%, -100 mesh) and hafnium (99.9%, -100 mesh) were obtained from Beijing Metallurgy and Materials Technology Co., China. nickel (99.99%, -100 mesh), iron (99.9%, -100 mesh), iodine (99.9%, granules), $SeCl_4$ (99.5%), WCl_6 (99.9%), $TeCl_4$ (99.9%), cobalt (99.9%, -100 mesh) were obtained from Strem, USA, platinum (99.99%, -100 mesh) was obtained from SurePure, USA. Palladium (99.95%, -100 mesh) was obtained from Safina, Czech Republic.

MoS_2: MoS_2 of natural origin was collected in Krupka, Czech Republic by Z.S.

Commercial crystals: Bi_2O_2Se (2D semiconductors), SnS (2D semiconductors), SnSe (2D semiconductors), TlSe (2D semiconductors), tellurium (Novaelements) and germanium (Novaelements) were bought commercially.

Black Phosphorus: BP was made by transport of red phosphorus in presence of mineralizer using standard procedure with SnI_4/Sn. Red phosphorus (2 g) and Sn (30 mg)/SnI_4 (60 mg)

(mineralizing agent) were placed in a quartz glass ampule and subsequently sealed using an oxygen/hydrogen torch under high vacuum (under $1x10^{-3}$ Pa using oil diffusion pump and LN_2 trap). The ampule was placed horizontally in a muffle furnace and heated to 600 °C in 8 hours. After 6 hours on temperature the ampoule was cooled on 400°C over a period of 50 hours, subsequently on 200°C in 25 hours and finally freely cooled at room temperature for 2 h. The obtained BP crystals with a metallic sheen were washed with CS2 to remove the byproduct of white phosphorus and stored in argon glovebox.

GeS: GeS was made by direct reaction of elements (25g) in stochiometric ratio in quartz ampoule (100 × 25mm) sealed under high vacuum (under 1×10^{-3} Pa using oil diffusion pump and LN_2 trap). The ampoule were placed horizontally in muffle furnace and heated above melting point. The ampoule was heated using 0.5 °C min^{-1} on 700°C and after 6 hours cooled on room temperature sing 0.1°C min^{-1}. Ampoule was open in glovebox and sample was kept under argon till further use.

GeSe: GeSe was made by direct reaction of elements (25g) in stochiometric ratio in quartz ampoule (100 × 25mm) sealed under high vacuum (under $1x10^{-3}$ Pa using oil diffusion pump and LN_2 trap). The ampoule were placed horizontally in muffle furnace and heated above melting point. The ampoule was heated using 1°C min^{-1} on 700°C and after 6 hours cooled on room temperature sing 0.1°C min^{-1}. Ampoule was open in glovebox and sample was kept under argon till further use.

$SnSe_2$: $SnSe_2$ was made by direct reaction of elements (25g) in stochiometric ratio in quartz ampoule (100×25mm) sealed under high vacuum (under 1×10^{-3} Pa using oil diffusion pump and LN_2 trap). The ampoule were placed horizontally in muffle furnace and heated above melting point. The ampoule was heated using 1°C min^{-1} on 700°C and after 6 hours cooled on room temperature sing 0.1°C min^{-1}. Ampoule was open in glovebox and sample was kept under argon till further use.

GaTe: GaTe was made by direct reaction of elements (25g) in stochiometric ratio in quartz ampoule (100×25mm) sealed under high vacuum (under 1×10^{-3} Pa using oil diffusion pump and LN_2 trap). The ampoule were placed horizontally in muffle furnace and heated above melting point. The ampoule was heated using 1°C min^{-1} on 830°C and after 6 hours cooled on

room temperature sing 0.1°C min^{-1}. Ampoule was open in glovebox and sample was kept under argon till further use.

InTe: InTe was made by direct reaction of elements (25g) in stochiometric ratio in quartz ampoule (100×25mm) sealed under high vacuum (under 1×10^{-3} Pa using oil diffusion pump and LN_2 trap). The ampoule were placed horizontally in muffle furnace and heated above melting point. The ampoule was heated using 1°C min^{-1} on 710°C and after 6 hours cooled on room temperature sing 0.1°C min^{-1}. Ampoule was open in glovebox and sample was kept under argon till further use.

InSe: InSe was made by direct reaction of elements (25g) in ratio 52 at.% In and 48 at.% Se in quartz ampoule (100×25mm) sealed under high vacuum (under 1×10^{-3} Pa using oil diffusion pump and LN_2 trap). The ampoule were placed horizontally in muffle furnace and heated above melting point. The ampoule was heated using 1°C min^{-1} on 710°C and after 6 hours cooled on room temperature sing 0.1°C min^{-1}. Ampoule was open in glovebox and sample was kept under argon till further use.

$PtSe_2$: $PtSe_2$ was made by direct reaction from elements in quartz glass ampoule of 100×25 mm dimension. For the synthesis of $PtSe_2$ was in ampoule placed 3g of Pt and Se with 2at.% excess of selenium. The ampoule was placed in muffle furnace and heated on 800°C for 25 hours and subsequently was heated on 1280°C (1°C min^{-1}) and after 1 hour was cooled over a period of 100 minutes on 1200°C and subsequently on room temperature using cooling rate 1°C min^{-1}.

$PdSe_2$: For the synthesis of $PdSe_2$ the elements corresponding to 3g of $PdSe_2$ were placed in stochiometric ratio in ampoule and melt sealed under high vacuum (under 1×10^{-3} Pa using oil diffusion pump and LN_2 trap). The ampoule was heated on 820 °C (1°C min^{-1}) and after 5 hours cooled on room temperature using cooling rate 0.1°C min^{-1}.

$MoSe_2$, WSe_2, $Mo_{0.5}W_{0.5}Se_2$: The crystals were made by chemical vapor transport in quartz ampoule. The elements corresponding to the 50g of compound were placed in quartz ampoule (50 × 250mm) together with 2 at.% excess of selenium and 0.5g of $SeCl_4$ and melt sealed under high vacuum (under 1×10^{-3} Pa using oil diffusion pump and LN_2 trap). The ampoule was placed in muffle furnace and heated on 500°C for 25 hours, on 600°C for 50 hours and finally on 800°C for 50 hours. Subsequently the ampoule were placed in two zone furnace and first the growth zone was heated on 1000°C and source zone on 800°C, after two days the thermal gradient was reversed and the source zone was heated on 950°C while the growth zone was

kept at 850 °C for 14 days. The ampoule was open under argon atmosphere in glovebox and keep under argon atmosphere till further use.

Nb doped $MoSe_2$ and WSe_2: The Nb doped materials were made by mixing the elements in ration $W_{0.97}Nb_{0.03}Se_2$ ($Mo_{0.97}Nb_{0.03}Se_2$) with total mass of 50g and 2 at.% excess of Se and 0.5g of $SeCl_4$ were placed in ampoule and melt sealed under high vacuum (under 1×10^{-3} Pa using oil diffusion pump and LN_2 trap). The ampoule was placed in muffle furnace and heated on 500°C for 25 hours, on 600°C for 50 hours and finally on 800°C for 50 hours. Subsequently the ampoule were placed in two zone furnace and first the growth zone was heated on 1000°C and source zone on 800°C, after two days the thermal gradient was reversed and the source zone was heated on 950°C while the growth zone was kept at 850 °C for 14 days. The ampoule was open under argon atmosphere in glovebox and keep under argon atmosphere till further use.

TaS_2: The crystals were made by chemical vapor transport in quartz ampoule. The elements corresponding to the 50g of compound were placed in quartz ampoule (50×250mm) together with 0.7g of I_2 and melt sealed under high vacuum (under 1×10^{-3} Pa using oil diffusion pump and LN_2 trap). The ampoule was placed in muffle furnace and heated on 500°C for 25 hours, on 600°C for 50 hours and finally on 800°C for 50 hours. Subsequently the ampoule were placed in two zone furnace and first the growth zone was heated on 1000°C and source zone on 800°C, after two days the thermal gradient was reversed and the source zone was heated on 950°C while the growth zone was kept at 850 °C for 14 days. The ampoule was open under argon atmosphere in glovebox and keep under argon atmosphere till further use.

$MoTe_2$: The crystals were made by chemical vapor transport in quartz ampoule. The elements corresponding to the 50g of compound were placed in quartz ampoule (50×250 mm) together with 2 at.% excess of tellurium and 0.5g of $TeCl_4$ and melt sealed under high vacuum (under 1×10^{-3} Pa using oil diffusion pump and LN_2 trap). The ampoule was placed in muffle furnace and heated on 500°C for 25 hours, on 600°C for 50 hours and finally on 800°C for 50 hours. Subsequently the ampoule were placed in two zone furnace and first the growth zone was heated on 1000°C and source zone on 800°C, after two days the thermal gradient was reversed and the source zone was heated on 900°C while the growth zone was kept at 800 °C for 14 days. The ampoule was open under argon atmosphere in glovebox and keep under argon atmosphere till further use.

WS_2: The crystals were made by chemical vapor transport in quartz ampoule. The elements corresponding to the 50g of compound were placed in quartz ampoule (50×250 mm) together with 2 at.% excess of sulfur and 0.5g of WCl_6 and melt sealed under high vacuum (under 1×10^{-3} Pa using oil diffusion pump and LN_2 trap). The ampoule was placed in muffle furnace and heated on 500°C for 25 hours, on 600°C for 50 hours and finally on 800°C for 50 hours. Subsequently the ampoule were placed in two zone furnace and first the growth zone was heated on 1000°C and source zone on 800°C, after two days the thermal gradient was reversed and the source zone was heated on 1000°C while the growth zone was kept at 900 °C for 14 days. The ampoule was open under argon atmosphere in glovebox and keep under argon atmosphere till further use.

$HfSe_3$: The crystals were made by chemical vapor transport in quartz ampoule. The elements corresponding to the 50g of compound were placed in quartz ampoule (50×250mm) together with 2 at.% excess of selenium and 0.5g of HfCl4 and melt sealed under high vacuum (under 1×10^{-3} Pa using oil diffusion pump and LN_2 trap). The ampoule was placed in muffle furnace and heated on 500°C for 25 hours, on 600°C for 50 hours and finally on 800°C for 50 hours. Subsequently the ampoule were placed in two zone furnace and first the growth zone was heated on 1000°C and source zone on 800°C, after two days the thermal gradient was reversed and the source zone was heated on 900°C while the growth zone was kept at 800 °C for 14 days. The ampoule was open under argon atmosphere in glovebox and keep under argon atmosphere till further use.

$NiPS_3$ and $FePS_3$: The crystals were made by chemical vapor transport in quartz ampoule. The elements corresponding to the 30g of compound were placed in quartz ampoule (50×250mm) together with 1 at.% excess of sulfur and phosphorus and 0.5g of I2 and melt sealed under high vacuum (under 1×10^{-3} Pa using oil diffusion pump and LN_2 trap). The ampoule was placed in muffle furnace and heated on 500°C for 25 hours, on 600°C for 50 hours and finally on 700°C for 50 hours (heating rate was 0.2°C min^{-1}). Subsequently the ampoule were placed in two zone furnace and first the growth zone was heated on 750°C and source zone on 600°C, after two days the thermal gradient was reversed and the source zone was heated on 750°C while the growth zone was kept at 650 °C for 14 days. The ampoule was open under argon atmosphere in glovebox and keep under argon atmosphere till further use.

$CoPS_3$: The crystals were made by chemical vapor transport in quartz ampoule. The elements corresponding to the 15g of compound were placed in quartz ampoule (35×150mm) together with 1 at.% excess of sulfur and phosphorus and 0.5g of I_2 and melt sealed under high vacuum (under 1×10^{-3} Pa using oil diffusion pump and LN_2 trap). The ampoule was placed in muffle furnace and heated on 560°C for 30 days (heating rate was 0.1°C min^{-1}). The ampoule was open under argon atmosphere in glovebox and keep under argon atmosphere till further use.

Data Availability: Relevant data supporting the key findings of this study are available within the article and the Supplementary Information file. All raw data generated during the current study are available from the corresponding authors upon request.

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Author Contributions: T.C. conceived and designed the experiments. T.C. wrote the paper in close consultation with other authors. T.C. exfoliated the crystals, manufactured the inks and transistors. T.C. undertook I-V measurements, AFM, Raman spectroscopy and optical microscopy. K.S. made LS networks with T.C. and J.N. The DFT simulations were undertaken by N.M. and C.L. Growth of the 2D crystals was completed by Z.S, J.P. and I.P.. R.W and M.MC undertook XRD of the crystals. S.L undertook the UV-vis for the electronic inks. T.C. undertook AFM and A.D. completed the formal analysis. E.C did SEM and measured the TaS_2 stretchable conductor. G.G. and L.D.A.S. undertook the THz spectroscopy measurements of the networks. J.C. and E.C. implemented impedance model. L.A. and R.S. fabricated/implemented the circuits and performed the measurements. T.C. and J.C. acquired funding for the project and wrote the paper.

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Figure 1: Material families and mechanical criteria for exfoliation. a Schematic of the systematic down selection process for identifying low-bandgap 2D materials for solution-processed circuits. **b** Optical images of various crystals examined in the study, highlighting different families of 2D materials with corresponding color-coded outlines with transition metal trichalcogenides (TMT, green), transition metal dichalcogenides (TMD, orange), transition metal monochalcogenides (TMM, blue), elemental materials (red), and bismuth oxychalcogenides (BCT, yellow). **c** Plot of in-plane (E_{in}) versus out-of-plane (E_{out}) Young's moduli for each material, with a dashed line indicating $E_{in}/E_{out} = 1$. **d** Correlation between simulated mechanical properties (E_{in}/E_{out} and interlayer binding energy, E_b) and experimentally determined intercalation success. **e** Semiconducting and metallic inks after dispersion in N,N dimethylformamide with polyvinylpyrrolidone (DMF-PVP).

Figure 2: Nanosheet dimensions and formation of solution processed networks. a Atomic force microscopy (AFM) micrograph showing InSe nanosheets drop-cast onto a Si/SiO_2 substrate. **b-c** Nanosheet lateral size , *L*, (**b**) and aspect ratio, *AR*, (**c**) plotted versus apparent nanosheet thickness , *t*, for four selected nanosheet types. Each data point represents a single nanosheet. **d** Relationship between average nanosheet lateral size (<*L*>) and apparent thickness (<*t*>) for each material under study. In **d**, the green region illustrates $E_{in}/E_{out}>1.7$. The data has been grouped into families of materials: TMT, TMD, TMM, and elemental materials. **e** Dependence of mean nanosheet *AR* for each material on the starting crystal's E_{in}/E_{out} ratio. The data has been grouped into material families, as shown in **d**. Errors in **d** and **e** are the standard

deviation of the mean, SDOM. **f** Scanning electron microscopy images showing the morphology of solution-deposited nanosheet networks prepared using nanosheets from each material family. These images show alignment and connectivity for high *AR* nanosheets and the presence of nanosheets in the tellurene network. **g** Raman spectra of selected 2D materials showing characteristic vibrational modes, indicating the phase and quality of exfoliated nanosheets.

Figure 3: Conductivity and electrical behaviour of nanosheet networks. a Optical images showing a nanosheet network fabricated via Langmuir–Schaefer (LS) coating on polyethylene terephthalate (PET) substrates pre-patterned with gold contacts for source, drain, and gate electrodes. Channel width (W_{ch}) and channel length (L_{ch}) are indicated. **b** Network conductivity (σ_{Net}) plotted as a function of optical bandgap (E_{Op}) for different material families. The horizontal dashed line indicates the conductivity threshold below which networks could not be electrochemically gated. **c** Fractional resistance change ($\Delta R/R_0$) plotted versus strain for a TaS_2 network, demonstrating linear scaling with very small slope. The gauge factor (GF), defined as $\Delta R/R_0/\varepsilon$ where ε is strain, is low ~ 1, indicating near strain-independent resistance. Inset: The resistance is almost invariant to a sawtooth applied strain (varying from 0% to 2% to 0% at 0.5 Hz). **d** Schematic of the flexible electrochemically gated transistor on PET. The device consists of source (S), drain (D), and gate (G) electrodes. A drain–source voltage (V_{ds}) is applied between the source and drain, while a gate voltage (V_{gs}) is applied to the gate to modulate the channel conductivity. **e-g** Electrochemically switched transfer characteristics of various networks indicating n-type (**e**), ambipolar (**f**) and p-type (**g**) behaviour. **h-i** Summary of the performance of electrochemically gated solution-processed transistors studied in this work, showing comparison of network mobility (μ_{Net}) and on/off current ratio (I_{on}/I_{off}) (**h**) as well as network conductivity (σ_{Net}) and charge carrier density (n) for each network (**i**). Not shown in f are data for GaTe and InTe networks due to their extremely small mobility ($<10^{-7}$ cm^2 V^{-1} s^{-1}) and on/off ratio (<2).

Figure 4: Nanosheet and network mobility with junction resistance analysis. a Nanosheet mobility ($\mu_{NS}(\omega)$) found from THz spectroscopy as a function of radian frequency ($\omega/2\pi$) of the THz electric field for a range of different nanosheet types. **b** Nanosheet mobility (μ_{NS}) found from THz spectroscopy plotted as a function of network mobility (μ_{Net}) shown on Figure 3h. Errors are found by SDOM over 5 independent devices. **c** Real impedance spectra measured on TMD networks. These spectra are scaled to represent the real impedance of single (average)

nanosheet-junction pairs. The solid lines represent fits to an equivalent circuit model. **d** The junction (R_J) and nanosheet resistance (R_{NS}) were extracted from the data in **c** for each material examined. Errors are found by SDOM over 5 independent devices. **e** The relation between R_J/R_{NS} ratio and the ratio of nanosheet to network mobility (μ_{NS}/μ_{Net}). The dashed line represents the expected behaviour (see text)**.** Error bars are found by fractional propagation of errors. **f** The ratio of μ_{Net}/μ_{NS} plotted versus mean aspect ratio. The dashed line is a guide to the eye. Errors are found by SDOM over 5 independent devices combined with error propagation.

Figure 5: **Solution processed DAC and BASK circuits. a** Bright-field optical microscopy image of the WS_2 digital-to-analog converters. **b** Magnified optical image of the solution-processed WS_2 four-bit digital-to-analog converter (DAC) with the corresponding circuit diagram. The DAC is based on a binary weighted resistor network. The outputs are referenced to ground (GND). The input lines are labelled a_0, a_1, a_2, a_3 for the resistor connections and b_0, b_1, b_2, b_3 for the corresponding digital bit inputs, where b_3 is the most significant bit and b_0 the least significant bit. **c** Circuit diagram of the analog-to-digital converter (ADC) connected with three bits to the solution-processed DAC, together with the corresponding logic table. The input analog voltage (V_{IN}) is converted by the ADC into a digital signal, which is then supplied to the DAC. The DAC produces a reconstructed analog output voltage (V_{OUT}). The digital outputs of the ADC are referenced to the supply voltage (V_{DD}). **d** Digital signals from bit lines in a 3-bit DAC, showing the contribution of each digital signal to the output voltage. **e** Analog input to the ADC (orange line) and the corresponding analog output (purple line) from the 3-bit WS_2 DAC. **f** Circuit diagram of the binary amplitude shift keying (BASK) circuit used for signal encoding. A high-frequency local oscillator with frequency (f_{LO}) provides a sinusoidal carrier signal with amplitude denoted as V_{LO}. The digital input controls the modulation of the carrier, producing an amplitude-encoded output. **g** Digital signal encoding into a high-frequency analog signal. The digital message input into the top FET gate modulates the high-frequency carrier signal to produce the encoded output. **h** Binary bits required to spell the word Dublin in 7-bit ASCII. V_{IN} represents the digital message shown by the blue curve. The output of the BASK circuit which has the encoded digital message in the local oscillator is shown by the orange curve and the decoded signal is shown as the purple line.