Transition from Quasi-2D to Quasi-1D van der Waals Materials:
Electronic Properties of TaSe$_3$/h-BN Heterostructures

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A charge-density-wave oscillator based on an integrated tantalum disulfide-boron nitride-graphene device operating at room temperature

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The $V_{DC}$ bias is applied at the drain terminal of the G-FET and the $V_G$ bias is connected to the gate terminal of G-FET. Ground is connected to one terminal of 1T-TaS$_2$ device, while the common terminal of the two devices is the output port.

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Charge Density Waves: Basics

Normal state $T > T_c$

Electron Density $\rho_e = \text{const}$

Atomic Spacing $\frac{\pi}{a}$

Energy

Peierls state $T < T_c$

Electron Density $\rho_e = \rho_0 + \rho_1 \cos(qx + \lambda_c)$

Atomic Spacing

Energy


The threshold switching (TS) effect is prominent from 78 K to 320 K. The blues arrows indicate the voltage sweep direction for the measurement at 78 K. For all the other temperatures, $V_H$ is always higher than $V_L$. The hysteresis window is defined as $V_H - V_L$. The TS is prominent up to 320 K, and becomes less pronounced as the temperature approaches the NC-CDW–IC-CDW transition at 350 K. As shown in the inset, at 345 K (red curve), the TS is still measurable. As T exceeds 350 K, the IV becomes linear.
An Integrated 1T-TaS$_2$ – h-BN – Graphene Oscillator

The SEM image of the integrated 1T-TaS$_2$–BN–graphene voltage controlled oscillator.

Output waveforms at different gate biases when $V_{DC}$ is fixed at 3.65 V. The oscillation frequency is tunable with gate biases in the range of 0.68 V to 1.8 V. The different waveforms are vertically offset of 0.25 V for clarity.

Fundamental Science Motivation: From Quasi-2D to Quasi-1D van der Waals Materials

Can we exfoliate Quasi-1D atomic threads like we do quasi-2D atomic planes?

MoS$_2$

\[ \rightarrow \text{Crystal structure of monoclinic TaSe}_3, \text{ with alternating layers of TaSe}_3 \]

\[ \rightarrow \text{Cross section of the unit cell, perpendicular to the chain axis (b axis).} \]

\[ \rightarrow \text{The side view: 1D nature of TaSe}_3 \text{ chains along the b axis.} \]
Quasi-1D Crystals Can be Mechanically and Chemically Exfoliated from Bulk TaSe$_3$

High resolution scanning transmission electron microscopy image of exfoliated TaSe$_3$ showing pristine metal trichalcogenide chains that extend along the b axis.

Scanning electron microscopy image of TaSe$_3$ crystals prepared by CVT method.

Collaboration:
Prof. Tina T. Salguero
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Chemical Vapor Transport (CVT) Method
Crystallinity of Quasi-1D TaSe$_3$ Metal

(b) SEM image of a TaSe$_3$ crystal used in this work. (c) HRTEM image of TaSe$_3$ after solvent exfoliation. The inset shows the position of the observed (1 0 -1) lattice plane in the van der Waals gap. (d) Powder XRD pattern of TaSe$_3$ crystals; the experimental data in black matches the reference pattern in blue (JCPDS 04-007-1143). The intensities of peaks marked with * are enhanced due to orientation effects. (e) Raman spectrum of TaSe$_3$ threads under 633 nm laser excitation.
Selective area electron diffraction data for five various locations along the length of the TaSe$_3$ nanowires confirming the crystallinity and uniformity of the sample.
Practical Motivations for Quasi-1D Metals: Search of New Interconnect Materials

According to ITRS:

- Current density $\sim 1.8$ MA/cm$^2$ at the half-pitch width of 28.5 nm will increase to $\sim 5.35$ MA/cm$^2$ at the width of 7 nm.

- There is no existing technology with the breakdown current density high enough to sustain such currents.

- The layer thicknesses will decrease from 57.0 nm in 2016 to 15.4 nm by 2028.
Boron Nitride Capped Devices with Quasi-1D TaSe$_3$ Channels

Boron Nitride capping is essential for device fabrication.

Schematic of the TaSe$_3$/h-BN quasi-1D / quasi-2D nanowire heterostructures used for the I-V testing.

The metals tested for fabrication of Ohmic contacts included combinations of thin layers of Cr, Ti, Au, Pd together with a thicker Au layer.
Quasi-1D TaSe$_3$ Nanowires: Surface Roughness
Low-Field Electrical Characteristics of Devices with Quasi-1D TaSe$_3$ Channels

→ Current-voltage characteristics of TaSe$_3$ devices with different channel length.

→ Linear characteristics at low voltage indicates good Ohmic contact of TaSe$_3$ channel with the metal electrodes.

The contact resistance extracted from TLM data is $2R_C = 22 \ \Omega \cdot \mu$m
Current Density in Quasi-1D TaSe$_3$ Nanowires

→ High-field I-V characteristics showing the breakdown point. In this specific device the breakdown is gradual.

→ Breakdown current density of about 32 MA/cm$^2$ — an order-of-magnitude higher than that for copper.

Step-Like Breakdown in Quasi-1D TaSe$_3$

(a) Current-voltage characteristics of a device with Cr/Au (10/150 nm) contacts. Note a step-like breakdown starting at $J_B = 4 \times 10^6$ A/cm$^2$. (b) Current-voltage characteristics of devices with pure Au contacts (150 nm) showing the step-like breakdowns at $J_B = 6.1 \times 10^6$ A/cm$^2$ (black), $5.7 \times 10^6$ A/cm$^2$ (blue), and $6.3 \times 10^6$ A/cm$^2$ (red).
Pulse Measurements of TaSe$_3$ Nanowires

Breakdown mechanism: thermal vs. electromigration

Duration and shape of the pulses applied to the h-BN / TaSe$_3$ nanowire devices (left). I-V characteristics of the devices measured in the pulse and DC regimes (right).

Conclusion: self-heating does not play a major role.
The Fuchs-Sondheimer model for the electron–nanowire surface scattering and the Mayadas-Shatzkes model for the electron–grain boundary scattering give the following expression for electrical resistivity:

\[
\rho = \rho_0 \left[ 2C\lambda_0 \cdot (1 - p) \cdot \left( \frac{1}{H} + \frac{1}{W} \right) + \frac{1}{1 - 3\alpha / 2 + 3\alpha^2 - 3\alpha^3 \ln(1 + 1/\alpha)} \right]
\]

\[\alpha = \lambda_0 \cdot R/(d_G \cdot (1 - R))\]

- \(\rho_0\) - bulk electrical resistivity
- \(W\) - nanowire width
- \(H\) - nanowire thickness
- \(\lambda_0\) - bulk electron MFP
- \(C = 1.2\) – shape factor
- \(R\) - reflectivity parameter of the electron – grain boundary scattering
- \(p\) – specularity parameter for electron – surface scattering
- \(d_G\) - average grain size

The first term describes the electron scattering on nanowire surface roughness while the second term corresponds to the electron scattering on grains.

Comparison with Copper Interconnects

Calculated electrical resistivity of Cu nanowires normalized to the bulk Cu resistivity as a function of the nanowire width $W$. The results are shown for a range of specularity parameters $p$, which defines electron scattering from nanowire surfaces and parameter $R$, which determines the electron scattering from grain boundaries.

The increase in TaSe$_3$ resistivity is expected to be less drastic owing to the absence of grain boundaries ($R \rightarrow 0$) and smoother surfaces ($p \rightarrow 1$).
Low-Frequency Flicker Noise

The noise measurement set-up is placed inside a special room with the metal and acoustic protection from the environmental noises and electro-magnetic fields.

Low noise batteries are used for the biasing of the devices.
Low-Frequency Noise as Reliability Metric

Noise spectrum of TaSe$_3$ devices at room temperature. The noise spectrum $S_I$ is flowing $1/f^\gamma$ dependence with $\gamma = 1.1\sim 1.2$. The inset shows the noise level at $f = 10$ Hz as the function of the channel current.

The quadratic dependence of the noise spectrum density $S_I$ on the channel current $I$ indicates that the $1/f$ noise measured at this current level originates from the TaSe$_3$ device itself rather than the current induced effects.

Comparison with Graphene and CNTs

Normalized noise spectrum density as a function of the resistance for different low-dimensional material systems - quasi-1D TaSe$_3$ nanowires, graphene and carbon nanotubes. For comparison, the empirical relation $A=10^{-11} R$ for the low-frequency noise versus resistance $R$.

The 1/f noise at RT becomes more of $1/f^2$ type at elevated temperatures. The increased frequency power factor $\gamma$ suggests the onset of the electromigration processes.

Inset shows the temperature dependent resistance of the quasi-1D TaSe$_3$ nanowire.

The graduate increase of the resistance with temperature, for $T < 410$ K is typical for metal. The sharply rising resistance for $T > 410$ K indicates the occurrence of electromigration.
The frequency power factor $\gamma$ increases from 1.16 - 1.24 at 298 K to 1.72 - 1.75 at 350 K, and remains approximately constant at 1.71-1.75 above 350 K. The blue curve is the fitting of the experimental data.

The correlation of the frequency power factor $\gamma$ with the noise (electromigration) activation energy $E_P$ is given by:

$$\gamma(T) = 1 - \frac{1}{\ln(2\pi f \tau_0)} \left( \frac{\partial \ln S(f, T)}{\partial \ln T} - 1 \right)$$

$$S(f, T) \propto \frac{kT}{2\pi f} D(E)$$

$$E_P = -kT \ln(2\pi f \tau_0)$$
Temperature dependent $1/f^2$ noise analysis using the Arrhenius plot of $T \times S/I^2$ versus $1000/T$. The extracted electromigration activation energy for quasi-1D TaSe$_3$ nanowire is $E_A = 0.88$ eV.

Conclusions

- The concept and approaches of quasi-2D van der Waals materials can be extended to quasi-1D van der Waals materials.
- Demonstrated record high 32 MA/cm$^2$ current density in a few devices and on average above 10 MA/cm$^2$ in numerous devices.
- Quasi-1D van der Waals metals allow for ultimate scaling limit – individual atomic thread (1 nm $\times$ 1 nm).
- Resistivity scaling in quasi-1D metals can be slower than for conventional metals as the nanowire cross-section decreases.
- Breakdown is likely of electromigration nature.
- It is possible that other quasi-1D metals may have lower resistivity.