Low-Frequency Noise in Low-Dimensional van der Waals Materials

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Graphene, Phonons and Thermal Properties

Areas of Expertise

2D/1D Materials and Devices, and Low-Frequency Noise

Low-frequency $1/f$ noise in graphene devices

Alexander A. Balandin

Low-frequency noise with a spectral density that depends inversely on frequency has been observed in a wide variety of systems including current fluctuations in resistors, intensity fluctuations in music and signals in human cognition. In electronics, the phenomenon, which is known as $1/f$ noise, flicker noise or excess noise, hampers the operation of numerous devices and circuits, and can be a significant impediment to the development of practical applications from new materials. Graphene offers unique opportunities for studying $1/f$ noise because of its two-dimensional structure and widely tunable two-dimensional carrier concentration. The creation of practical graphene-based devices will also depend on our ability to understand and control the low-frequency noise in this material system. Here, the characteristic features of $1/f$ noise in graphene and few-layer graphene are reviewed, and the implications of such noise for the development of graphene-based electronics including high-frequency devices and sensors are examined.
Outline

→ Introduction

→ Noise Experiments with Graphene
  → Graphene under irradiation
  → Noise as signal in graphene sensors

→ Noise in 2D CDW Devices
  → 2D charge-density-waves
  → Noise spectroscopy of CDWs
  → Radiation hardness of 2D CDW devices

→ Quasi-1D van der Waals Materials
  → Interconnect applications of 1D metals
  → Unusual noise characteristics in 1D

→ Conclusions
Terminology: Van der Waals Materials

From quasi-2D to quasi-1D

Graphene

MoS$_2$

TaSe$_3$

Crystal structure of monoclinic TaSe$_3$, with alternating layers of TaSe$_3$

Cross section of the unit cell, perpendicular to the chain axis (b axis).

The side view: 1D nature of TaSe$_3$ chains along the b axis.
Part – I: Noise in Graphene

→ Practical task of noise scaling with the thickness

→ Possibility of addressing an old problem of origin of noise: surface vs. volume

The back-gated devices were fabricated by the electron-beam lithography with Ti/Au (6-nm/60-nm) electrodes.

\[ R_{ST} \] is sheet resistance

Graphene channel area \( A \) varied from 1.5 to 70 \( \mu m^2 \)
Electronic Noise in Graphene Devices

To minimize the influence of the contact resistance, most of the noise measurements were performed close to CNP.

Old Question: Surface vs. Volume Effect

The intensity of discussions can be inferred even from the titles of seminal publications on the subject: “1/f noise is no surface effect” (1969) followed by “1/f noise: still a surface effect” (1972).

- 1/f noise is dominated by the volume noise when the thickness exceeds ~7 atomic layers (~2.5 nm).
- The 1/f noise is the surface phenomenon below this thickness.
Mechanism of $1/f$ Noise in Electronic Materials and Devices

$I \sim qN\mu$

$\delta I \sim q(\delta N)\mu + qN(\delta \mu)$

McWhorter’s model:

$g(\tau_N) = \left[\tau_N \ln(\tau_2 / \tau_1)\right]^{-1}$

$S_N(\omega) = 4\delta N^2 \int_{\tau_1}^{\tau_2} g(\tau_N) \frac{\tau_N}{1 + (\omega \tau_N)^2} d\tau_N$
Specifics of Electronic Noise in Graphene

\[ A = \frac{1}{N} \sum_{m=1}^{N} f_m S_{I_m} / I_m^2 \]

In some graphene devices, V-shape becomes M-shape dependence at larger bias range.

\[ S/I^2 = 10^{-9} \text{ to } 10^{-7} \text{ Hz}^{-1} \text{ at } f=10 \text{ Hz or } A=10^{-9} - 10^{-7} \]

\[ (S/I^2) L \times W = 10^{-8} - 10^{-7} \mu m^2/Hz \]
Investigation of $1/f$ Noise in Graphene Devices under Irradiation

**Goal**

Controlled introduction of defects by electron beam irradiation and observation of the evolution of $1/f$ noise level

**Methodology**

- **Step I:** Raman of pristine graphene
- **Step II:** IV characteristics and noise measurements
- **Step III:** E-beam irradiation of the device
- **Step IV:** Raman of the irradiated device
- **Step V:** IV characteristics and noise measurements

![Diagram of a graphene device with metal contacts, source, drain, and Si/SiO$_2$ substrate]
Introduction of Structural Defects to Graphene by Electron Beam Irradiation

The electron energy was set to 20 keV in order to exclude the severe knock-on damage to the graphene crystal lattice, which starts at ~50 keV.
Electron Beam Irradiation Effects on Electronic Properties of Graphene

The Dirac point shift to negative side was observed for most devices, although in a very few cases, we recorded a positive shift after some irradiation steps.
Electronic Noise Suppression via Electron Beam Irradiation

The noise was measured in the linear region of the drain bias keeping the source at a ground potential.

The flicker $1/f$ noise is usually associated with structural defects.

Introduction of defects by irradiation normally results in increased $1/f$ noise and reduced mobility.

Low-Frequency Noise Suppression via Electron Beam Irradiation

Noise reduces monotonically with the increasing $R_D$ for the entire range of negative gate-bias voltages, $V_G - V_D$. The same trend was observed for the positive gate bias.
Possible Mechanisms of the 1/f Noise and its Suppression in Graphene

McWhorter model of the number of carriers fluctuations:

\[ \frac{S_I}{I^2} = \frac{\lambda kT N_t}{f A V n^2} \]

- \( N_t \) is the concentration of the traps near the \( E_F \) responsible for noise
- \( A \) is the gate area
- \( n \) is the carriers concentrations
- \( \lambda \) is the tunneling constant


\( N_t \) is not the total concentration of traps!

\( \rightarrow \) Reduction in \( N_t \) after irradiation? – possible but unlikely

\[ g(\tau_N) = \left[ \tau_N \ln(\tau_2 / \tau_1) \right]^{-1} \]

\[ S_N(\omega) = 4\delta N^2 \int_{\tau_1}^{\tau_2} g(\tau_N) \frac{\tau_N}{1 + (\omega \tau_N)^2} d\tau_N \]
Possible Mechanisms of the 1/f Noise in Graphene

Noise spectral density of the elementary fluctuation in the mobility fluctuation model:

\[
\frac{S_I}{I^2} \propto \frac{N_t^\mu}{V} \frac{\tau \zeta(1 - \zeta)}{1 + (\omega \tau)^2} l_0^2 (\sigma_2 - \sigma_1)^2
\]

- \( N_{t\mu} \) is concentration of the scattering centers contributing to 1/f noise
- \( l_0 \) is MFP of the charge carriers
- \( \zeta \) is the probability for a scattering center to be in the state with the cross-section \( \sigma_1 \)

\[ \Rightarrow \quad N_{t\mu} \text{ may change during the irradiation or may stay about the same} \]

\[ \Rightarrow \quad N_t \text{ that limit electron mobility increases} \]

\[ \Rightarrow \quad \text{Noise is defined by the electron MFP: } S/\mu I^2 (\sim l_0^2) \]

\[ \Rightarrow \quad \text{Reduced mobility results in reduced MFP} \]

\[ \Rightarrow \quad \text{In graphene } \mu \text{ is limited by the scattering from charged defects even at RT} \]


Independent Confirmation:

Low-Frequency Fluctuations as a Signal Rather than Noise

- High gas sensitivity (<1 ppb); Linear response to the gas concentration
- Sensing parameters: change in resistivity; shift in Dirac point voltage; frequency of SAW


Look for G-R bulges, which can be informative → sensor selectivity
Generation – Recombination Bulges in Spectrum of Graphene under Gas Exposure

- Vapors generated by bubbling dry carrier gas - air – through solvent and diluting the gas flow with dry carrier gas
- Vapors generated at ~0.5 \( \frac{P}{P_o} \), where \( P \) during the experiment and \( P_o \) is the saturated vapor pressure
- Measurements performed at \( V_G = 0 \) V ("hole" region of graphene I-V)
- The contact resistance per unit width: 0.2 – 2.0 \( \Omega \)-mm

Superposition of a Lorentzian noise with defined relaxation time on a 1/f noise results in a bulge with specific frequency

\[
S \propto \frac{1}{1 + (\omega \tau)^2}
\]
Reproducibility of G-R Bulges in Spectrum of Graphene under Vapor and Gas Exposure

→ The characteristics frequency $f_c$ of Lorentzian peaks with certain vapor gases is reproducible for different graphene devices.

→ The gas molecules can create specific traps and scattering centers in graphene, which lead to either number of carriers fluctuation due to the fluctuations of traps occupancy or to the mobility fluctuations due to fluctuations of the scattering cross sections.

Part – II: Noise of Charge Density Waves

Normal state $T > T_c$

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

Atomic Spacing \( \frac{a}{a} \)

Peierls state $T < T_c$

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

Atomic Spacing


CDW Effects in $1T$-$TaS_2$

1. $1T$-$TaS_2$ experiences several CDW transitions.
2. CDW phase transition is accompanied by an abrupt resistance change.
3. C-CDW to NC-CDW transition ($\sim 180$ K) appears in thick film but absent in thin films ($H<9$ nm).
4. NC-CDW to IC-CDW phase transition at 350-360 K is preserved as the thickness reduced.


2D Charge-Density-Wave Devices

Channel thickness:  
\[ t = 6 \text{ nm} - 9 \text{ nm} \]

Contacts:  
Pd/Au (15 nm / 60 nm)

→ The h-BN cap provides air stable passivation for the 1T-TaS$_2$.

→ The edge contacts provide good Ohmic contacts to the 1T-TaS$_2$.

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 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.
Low-Voltage I-V Characteristics of Thin Film 1T-TaS₂

→ Temperature-dependent resistance measurements for 1T-TaS₂. The NC-CDW–IC-CDW and IC-CDW–NC-CDW transitions happen at 350 K and 340 K during the heating and cooling process, respectively. The resistance is measured at low voltage (V=20 mV).

→ Resistances match

No transition at 180 K
Oscillator Based on 1T-TaS\textsubscript{2} Device

\[ V_{\text{DC}} \]
\[ R_s \]
\[ 1T-\text{TaS}_2 \]
\[ V_{\text{OUT}} \]
\[ C_\Sigma \]

\[ V_{\text{DC}} = 3.98V \]
\[ V_{\text{DC}} = 3.95V \]
\[ V_{\text{DC}} = 3.86V \]
\[ V_{\text{DC}} = 3.83V \]
\[ V_{\text{DC}} = 3.80V \]

- Circuit schematic of the oscillator consists of the 1T-TaS\textsubscript{2} film, a series connected load resistor, and a lumped capacitance from the output node to ground. The load resistance is 1 k\( \Omega \).

- The output terminal is monitored by an oscilloscope.

- Voltage oscillations under different \( V_{\text{DC}} \): The circuit oscillates when \( V_{\text{DC}} \) is within the range of 3.83-3.95 V. The frequency is 1.77 MHz, 1.85 MHz, and 2 MHz when \( V_{\text{DC}} \) is 3.83, 3.86 and 3.95 V, respectively.

Different operation mechanism from early devices — no de-pinning

Allows for high T operation
An Integrated $1T$-TaS$_2$ – h-BN – Graphene Oscillator

The device structure of the integrated oscillator consists of a graphene FET series connected to $1T$-TaS$_2$.

Both the graphene and TaS$_2$ are fully covered with h-BN which acts as a protection layer against oxidation for the $1T$-TaS$_2$ and as a gate dielectric for the G-FET.

The equivalent circuit is shown in the inset. 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.

An Integrated 1T-TaS$_2$ – h-BN – Graphene Oscillator

The SEM image of the integrated 1T-TaS$_2$–BN–graphene voltage controlled oscillator. The graphene and the TaS$_2$ are highlighted by dashed lines.

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.

An Integrated $1T$-TaS$_2$ – h-BN – Graphene Oscillator

The dependence of oscillation frequency as function of gate bias.

Blue circles show the frequency of the oscillation under increased gate bias. The frequency can be adjusted monotonically with the tuning sensitivity of 0.3 MHz/V.

The red squares are the resistance value of the G-FET under different gate biases with fixed $V_{DC}$=2.4V.
Circuits Based on CDW Devices

(a) Schematic of the inverter gate consisting of two CDW devices and one regular resistor. (b) Phase space of the circuit comprising two CDW devices. (c) Results of numerical modeling showing the output voltage $V_{out}$ as a function of $V_{in}$. (d) The change in the CDW resistances $R_1$ and $R_2$ as a function of $V_{in}$.

1T-TaS$_2$ CDW Devices Under X-Ray Irradiation

TID response of 1T-TaS$_2$ devices up to 1 M rad (SiO$_2$). (a) I-V curves measured after each X-ray irradiation step. (b) Threshold voltages, $V_H$ and $V_L$, threshold currents, $I_H$ and $I_L$ as function of dose. (c) Extracted resistance at the high resistance and low resistance states as a function of dose.

Carrier concentration:
$10^{21}$ cm$^{-2}$ - $10^{22}$ cm$^{-2}$

Radiation Hardness of CDW Devices

(a) Circuit schematic diagram of a self-sustaining oscillator implemented with one 1T-TaS$_2$ device and a load resistor. (b) Oscillation waveform before and after 1 Mrad(SiO$_2$) X-ray irradiation.

Proton Effect on 1T-TaS$_2$ Devices

The quasi-two-dimensional (2D) 1T-TaS$_2$ channels show a remarkable immunity to bombardment with the high-energy 1.8 MeV protons to, at least, the irradiation fluence of $10^{14}$ H$^+$cm$^{-2}$.

Noise Spectroscopy of CDW Sliding and Transitions

Unusual Features of Lorentzian Peaks in Noise of CDWs

A voltage increase of only 120 mV results in a four orders-of-magnitude change in $f_c$.

This drastic change in $f_c$ with the bias is highly unusual for conventional materials, where a Lorentzian spectrum is associated G-R noise with $f_c$ independent from the bias.


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RTS Noise in CDW Devices

(a) The noise spectral density after onset of sliding at different $V_b$. The corner frequency increases with increasing $V_b$.

(b) (c) (d) Time-domain signals at $V_b$ and time scales. Note that a small increase of the bias results in a significant change in the noise. The amplitude of the pulses increases and number of fluctuators becomes larger. This is different from classical RTS noise in semiconductor devices.

IC CDW – Metal Transition in Quasi-2D CDW Materials

Optical image of a representative device (left panel) and a schematic of the device layered structure (right panel). The scale bar is 2 µm.

Noise Spectroscopy of CDW Transitions

- Resistance as a function of the applied electric field measured at room temperature.
- Normalized current noise spectral density as the function of frequency for several values of the electric field, which include the point of transition from the incommensurate CDW to the normal metallic phase.
- Normalized current noise spectral density, measured at f=10 Hz, as the function of the electric field.
Vertical CDW Devices

Part – III: 1D Van der Waals Materials

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

Crystal structure of monoclinic TaSe$_3$, with alternating layers of TaSe$_3$

Cross section of the unit cell, perpendicular to the chain axis (b axis).

The side view: 1D nature of TaSe$_3$ chains along the b axis.

MoS$_2$

TaSe$_3$
Practical Motivations for the Search of New Interconnect Materials

According to ITRS:

→ Current density ~1.8 MA/cm² at the half-pitch width of 28.5 nm will increase to ~5.35 MA/cm² 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

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.


Resistivity is $2.6 - 6.4 \times 10^{-4}$ Ω-cm.
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.
Noise Analysis: Dutta–Horn Model

Correlation of the frequency power factor $\gamma$ with the noise (electromigration) activation energy $E_P$

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

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

\[
E_P = -kT \ln(2\pi f \tau_0)
\]

Quasi-1D ZrTe$_3$ van der Waals Nanoribbons

Noise Spectroscopy of the ZrTe$_3$ Quasi-1D van der Waals Nanoribbon Devices

(a) Normalized noise spectral density, $S/I^2$, as a function of frequency of quasi-1D ZrTe$_3$ nanoribbon at temperatures from 85 K to 280 K.

(b) Normalized noise spectral density multiplied by frequency, $S/I^2 \times f$, as a function of frequency. Note an unusually strong dependence of the Lorentzian peak on the temperature (at bias 0.1 V) The position of characteristics frequency, $f_c$, is shown in broken line.

$$S_I/I^2 = 
\frac{4N_t}{Vn^2} \frac{\tau F(1 - F)}{1 + (\omega \tau)^2}
$$

$$\frac{1}{\tau} = \frac{1}{\tau_c} + \frac{1}{\tau_e}
$$

$$\tau_c = \frac{1}{\sigma v_T n} \quad \tau_e = \frac{1}{\sigma v_T N_c \exp\left(\frac{-E_0}{kT}\right)}$$

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Arrhenius plot of the characteristics frequency, \( \ln(f_c) \), as a function of the inverse of temperature, \( K^{-1} \) in quasi-1D ZrTe\(_3\) device. This plot was used to extract the activation energy.

The extracted trap activation energy is \( E_0 \approx 0.18 \) eV.

\[
\sigma = \sigma_0 \exp\left(-\frac{E_1}{kT}\right).
\]

The activation energy of the cross-section temperature dependence and level position can be easily extracted as \( E_1=0.144 \) eV and \( E_0=0.0456 \) eV. These data indicate that the activation energy of the characteristic frequencies of the G-R noise peaks is dominated by the activation energy of the capture cross-section.

The Poole-Frenkel Effect or NOT?

(a) Normalized noise spectral density, $S/I^2$, as a function of frequency of quasi-1D ZrTe$_3$ nanoribbon at the source – drain voltage ranging from 45 mV to 250 mV. Inset shows a schematics of potential barrier height between two states. (b) Normalized noise spectral density multiplied by frequency, $S/I^2 \times f$, as a function of frequency. Note an unusually strong dependence of the Lorentzian peak on the source – drain bias (at $T = 78$K).

The shifting of the Lorentzian peak in the noise spectrum with the applied electric field can be due to reducing the impurity barrier potential in high electric field:

$$\Delta E_{fp} = \left( \frac{q^3 F}{\pi \varepsilon_0 \varepsilon} \right).$$

We estimated that the electric filed required to shift this frequency three orders of magnitude is of the order 50 kV/cm. However, for the 2-mm length nanoribbon the average field in the sample does not exceed ~1.25 kV/cm.

$$F \left( \frac{\varphi_1}{x + d/2} - \frac{\varphi_2}{-x + d/2} \right) \frac{1}{\ln L/a}$$

L.D. Landau & E.M. Lifshitz
Electrodynamics of Continuous Media (1960)
Conclusions

→ Typical graphene transistors reveal rather low level of the low-frequency noise: $S/I^2 = 10^{-9}$ to $10^{-7}$ Hz$^{-1}$ at $f=10$ Hz or $A=10^{-9} – 10^{-7}$

→ It is possible to reduce $1/f$ noise via electron beam irradiation

→ Noise reduction after irradiation is better explained by the mobility fluctuation models

→ Low-frequency fluctuations can be used for selective sensing with graphene

→ Demonstrated current density of $10$ MA/cm$^2 – 30$ MA/cm$^2$ in TaSe$_3$ and up to $100$ MA/cm$^2$ in ZrTe$_3$ nanowires

→ Low-frequency noise measurement is a convenient technique to monitor phase transitions and de-pinning in CDW materials and devices

→ Low-frequency noise was used for extraction activation energies

→ G-R noise reveals unusual behavior in quasi-1D materials
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