Low-Frequency Electronic Noise in Quasi-1D TaSe₃ van der Waals Nanowires

Guanxiong Liu,† Sergey Rumyantsev,‡§ Matthew A. Bloodgood,‖ Tina T. Salguero,‖ Michael Shur,‡ and Alexander A. Balandin†,*

†Nano-Device Laboratory (NDL) and Phonon Optimized Engineered Materials (POEM) Center, Department of Electrical and Computer Engineering, University of California, Riverside, Riverside, California 92521, United States
‡Department of Electrical, Computer, and Systems Engineering, Center for Integrated Electronics, Rensselaer Polytechnic Institute, Troy, New York 12180, United States
§Ioffe Institute, St. Petersburg 194021, Russia
‖Department of Chemistry, University of Georgia, Athens, Georgia 30602, United States

ABSTRACT: We report results of investigation of the low-frequency electronic excess noise in quasi-1D nanowires of TaSe₃ capped with quasi-2D h-BN layers. Semimetallic TaSe₃ is a quasi-1D van der Waals material with exceptionally high breakdown current density. It was found that TaSe₃ nanowires have lower levels of the normalized noise spectral density, S/I², compared to carbon nanotubes and graphene (I is the current). The temperature-dependent measurements revealed that the low-frequency electronic 1/f noise becomes the 1/f² type as temperature increases to ∼400 K, suggesting the onset of electromigration (f is the frequency). Using the Dutta–Horn random fluctuation model of the electronic noise in metals, we determined that the noise activation energy for quasi-1D TaSe₃ nanowires is approximately Eₐ ≈ 1.0 eV. In the framework of the empirical noise model for metallic interconnects, the extracted activation energy, related to electromigration is Eₐ = 0.88 eV, consistent with that for Cu and Al interconnects. Our results shed light on the physical mechanism of low-frequency 1/f noise in quasi-1D van der Waals semimetals and suggest that such material systems have potential for ultimately downscaled local interconnect applications.

KEYWORDS: Quasi-1D, van der Waals materials, low-frequency noise, interconnects, reliability

The investigations of two-dimensional layers and heterostructures have revealed new physics and demonstrated promising applications.1-14 Starting with graphene,1-5 and spreading to a wide range of layered van der Waals materials,6-10 successful isolation of individual atomic layers from their respective bulk crystals by mechanical exfoliation led to the fast growing research activities in the 2D materials field. In contrast to the layered van der Waals materials that yield 2D crystals, materials such as TaSe₃ and TiS₃ yield the quasi-one-dimensional (1D) van der Waals crystal structures. These materials belong to the group of transition metal trichalcogenides MX₃ (where M = Mo, W, and other transition metals; X = S, Se, and Te). In the monoclinal crystal structure of TaSe₃, the trigonal prismatic TaSe₃ units form continuous chains extending along the b-axis and leading to fiber- and needle-like crystals with anisotropic semimetallic or metallic properties. The quasi-1D atomic threads are weakly bound in bundles by van der Waals forces. As a consequence, the mechanical exfoliation of the MX₃ crystals results not in 2D layers but rather in quasi-1D van der Waals nanowires. We have recently demonstrated quasi-1D TaSe₃ nanowires with the record high current density exceeding J₉ ≈ 30 MA/cm², which is an order of magnitude larger than that for Cu interconnects.18

In this Letter, we report on the excess low-frequency electronic noise in quasi-1D nanowires of TaSe₃ capped with quasi-2D h-BN layers. The h-BN capping was used as a surface passivation protecting from environmental exposure. The measurements were performed in the temperature range from 300 to 430 K in order to elucidate the physical mechanism of the low-frequency noise and determine the noise activation energies. The low-frequency noise is a ubiquitous phenomenon present in all kinds of electronic devices including field-effect transistors, diodes, and interconnects.19-21 The frequency dependence and temperature characteristics of the spectral density of the low-frequency noise have been used as reliability metrics for interconnects in Si complementary metal–oxide–semiconductor (CMOS) technology. The low-frequency noise data at elevated temperatures are directly correlated with the electromigration failure mechanisms in interconnects.22-24
has been suggested that the low-frequency noise at high temperatures originates from the vacancy migration along the grain boundaries.\textsuperscript{22,23} Hence, the noise can reveal information on the electromigration failure of interconnects and metal thin films. The noise activation energies extracted from the two commonly accepted physical models, the Dutta–Horn model\textsuperscript{21,24} and the empirical noise model in metals,\textsuperscript{22,26,27} have shown an excellent agreement with the activation energies obtained from the industry standard electromigration mean-time-to-failure (MTF) tests. These considerations explain additional practical motivations for investigation of the low-frequency noise in quasi-1D nanowires of TaSe\textsubscript{3}, which exhibit promise as ultimately downscaled local interconnects owing to their single-crystal atomic thread structure and exceptionally high breakdown current density.

The bulk TaSe\textsubscript{3} crystals were synthesized by the chemical vapor transport (CVT) method and then solvent and mechanically exfoliated (see Methods section). The high quality of both bulk and exfoliated quasi-1D TaSe\textsubscript{3} samples has been confirmed using a variety of experimental techniques including high-resolution transmission electron microscopy, Raman spectroscopy, energy dispersive spectroscopy, powder X-ray diffraction, and electron probe microanalysis, as reported previously.\textsuperscript{18} The devices for the low-frequency noise measurement were fabricated by the mechanical exfoliation of quasi-1D TaSe\textsubscript{3} nanowires on Si/SiO\textsubscript{2} substrates. We used the dry transferred h-BN flakes to protect the quasi-1D TaSe\textsubscript{3} nanowire channels from oxidation and chemical exposure during the fabrication processes. The devices were fabricated by standard e-beam lithography and e-beam metal evaporation.\textsuperscript{28} In order to make an electrical connection to the quasi-1D channels, we opened the contact area by dry etching the h-BN thin film and the underlying TaSe\textsubscript{3} nanowire while leaving the edge of quasi-1D channels in contact with the metal. Figure 1b shows a scanning electron microscopy (SEM) image of TaSe\textsubscript{3} crystals prepared by the CVT method. A high-resolution scanning transmission electron microscopy (STEM) inspection (Figure 1a) confirmed a single crystal nature of the material. Figure 1c is a SEM image of representative devices used for the current voltage ($I$–$V$) and electronic noise measurements.

Figure 2 presents typical $I$–$V$ characteristics of the devices with different channel lengths. The perfectly linear $I$–$V$s at low electric fields indicate a high-quality electrical contact between the metal electrode and the quasi-1D TaSe\textsubscript{3} nanowire. The h-BN capping was essential for achieving good Ohmic contacts between the metal electrodes and the TaSe\textsubscript{3} channel. The primary function of the h-BN capping layer was to prevent the oxidation during the fabrication, particularly before the final processing step of electrode metal deposition. Etching the h-BN capping before the last step of metal deposition ensured clean channel material for contacting with the electrode metal. Additional details of the h-BN capping and etching procedures have been reported by us elsewhere.\textsuperscript{17,18} The value of the contact resistance for a typical h-BN-capped quasi-1D TaSe\textsubscript{3} device, determined from the transmission line measurements, was $\approx 22 \Omega \mu m$.\textsuperscript{18} The inset to Figure 2 shows an optical image of a representative device used in this study. The light blue stripe between metal electrodes is the quasi-1D TaSe\textsubscript{3} nanowire while the green region is the h-BN capping layer. The brown background is the Si/SiO\textsubscript{2} substrate.

The low-frequency noise measurements were performed at both UCR and RPI using in-house built experimental setups. The equipment included a “quiet” battery, a potentiometer biasing circuit, a low noise amplifier, and a spectrum analyzer. The details of our noise measurement procedures have been reported elsewhere.\textsuperscript{28,29} Figure 3a shows a typical normalized noise spectral density $S(f)/f \propto 1/f^\gamma$ with $\gamma = 1.16$ (here $I$ is the channel source–drain current) in the TaSe\textsubscript{3} nanowires with 20 nm × 110 nm cross section. The measurements were performed at room temperature (RT). The noise spectral
The quadratic dependence of the noise spectrum density $S_f$ on the resistance for different low-dimensional material systems: quasi-1D TaSe3 nanowires, graphene, and carbon nanotubes. For comparison, the empirical relation $A = 10^{11}R$ for the low-frequency noise versus resistance $R$, derived from the carbon nanotube studies, is also shown. The noise in quasi-1D TaSe3 nanowires is about 1 order of magnitude lower than that in graphene.

To better understand the evolution of the low-frequency noise with temperature, we plotted the frequency exponent γ and the normalized noise spectral density, $S_f/R$, at high temperature. To determine the frequency exponent, the normalized noise spectral density was empirically obtained for the carbon nanotubes. Most of the tested carbon nanotube devices had a larger resistance and, as a result, a larger noise spectral density. The higher levels of $1/f$ noise typically are associated with a higher density of structural defects as well as mechanical and electromigration damage. One should keep in mind that the metal contact with carbon nanotubes or graphene is usually less Ohmic than that with TaSe3. It is likely that carbon nanotubes or graphene devices have substantially higher contact noise contribution than TaSe3 devices.

The resistivity of quasi-1D TaSe3 nanowires increases with temperature, similar to other metallic materials. The inset to Figure 4 shows the resistivity change in the temperature range from 298 to 450 K. The gradual 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.
representative devices with different channel cross-section areas at the frequency $f = 10$ Hz. The frequency exponent $\gamma$ 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. (b) Evolution of the normalized noise spectra density $S(f)$ with temperature $T$ at frequency 10 Hz. The blue curve is calculated from the Dutta–Horn model. The activation energy estimated from the Dutta–Horn model is $E_p = 1.01$ eV.

Figure 5. (a) Extracted frequency power factor $\gamma$ as the function of temperature. 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. (b) Evolution of the normalized noise spectra density $S(f)$ with temperature $T$ at frequency 10 Hz. The blue curve is calculated from the Dutta–Horn model. The activation energy estimated from the Dutta–Horn model is $E_p = 1.01$ eV.

The experimentally observed deviations from $\gamma = 1$ and temperature dependence of the noise indicate that $D(E)$ is not a constant. One should note here that the maximum in the $S(f, T)$ dependence on temperature does not always coincide with the maximum of $D(E)$ function. The temperature dependence of the frequency exponent $\gamma$ is expressed as

$$\gamma(T) = 1 - \frac{1}{\ln(2\pi f_0 T)} \left( \frac{\partial \ln S(f, T)}{\partial \ln T} - 1 \right)$$

(2)

Here $\tau_0$ is characteristic time constant referred to as the attempt escape time. In the original Dutta–Horn model, the time constant $\tau_0$ was assumed to be $\sim 10^{-14}$ s, which is the typical attempt escape time for random processes in solids. The exact value of $\tau_0$ does not strongly affect the low-frequency noise analysis. The blue curve in Figure 5a is a polynomial fit to the experimental data used in the further analysis.

In Figure 5b, the blue line is calculated using eq 2 with the approximated dependence of the frequency exponent $\gamma$ on temperature shown in Figure 5a. One can see that the Dutta–Horn model describes accurately the temperature dependence of the noise for $T < 380$ K. The saturation or even the decrease of noise at higher temperatures cannot be explained by this model. The latter is likely related to the onset of another low-frequency noise mechanism. For this reason, applying of the Dutta–Horn model for temperatures above $\sim 400$ K has to be done with caution. The activation energy $E_p$ at which the distribution function $D(E)$ attains its peak is related to temperature as

$$E_p = -kT \ln(2\pi \tau_0 f f_0)$$

(3)

The activation energy distribution calculated from the noise data in metals usually has a peak at $E_p \approx 1$ eV. Using the data for a representative device heated all the way to 430 K, we determined that the energy distribution reaches its maximum at $E_p \approx 1.0$ eV for TaSe$_3$ nanowires as well (data set for “Device 1” in Figure 5b). In a more conservative approach, increasing the temperature only to $\sim 390$ K, within the application domain of the Dutta–Horn model (data set for “Device 2” in Figure 5b), we determine the low bound of the position of the maximum of the activation energy distribution function $D(E)$. This value is somewhat smaller but still close to $\sim 1$ eV.

The noise spectral density $S(f) \sim 1/f^2$ with $\gamma \approx 2$ in metals is often attributed to the effects of electromigration. Although the detailed mechanism of the resistance fluctuations in metal films has not been established yet, many experiment indicate that the low-frequency noise, electromigration, and degradation in metals are related. The $1/f^2$ noise spectral density with $\gamma \geq 2$, which appears at high temperatures, has been widely used to extract the electromigration activation energy for interconnects. The frequency exponents as high as $\gamma > 4$ have been reported.

Attributed to the vacancy migration, $1/f^2$ noise has been proposed as a useful diagnostic tool. It was found that the low-frequency noise at high temperature follows the empirical equation

$$S(f) = \frac{A f^3}{T f^7} \exp\left(\frac{E_A}{kT}\right)$$

(4)
the $E_A$ values extracted from the low-frequency noise measurements are very close to those obtained from the conventional mean-time-to-failure test for interconnects fabricated with different technology and various metals, including Al, Al alloys, and Cu. The noise measurements have specific advantages over the conventional reliability tests, which require time-dependent resistance measurements of interconnects under high temperature and high current stress for long periods of time (typically from 10 to 100 h for each set). The noise measurements can be conducted in much shorter period of time. They are also be nondestructive. It was found that the noise level often increases rapidly before the sharp resistance increase, which was defined as the interconnect failure.24

Figure 6 shows the Arrhenius plot of $T \times S_i / f^2$ vs 1000/$T$ for $f = 10$ Hz. The extracted $E_A$ of 0.88 eV is within the range reported for other metal films. Table 1 summarizes the electromigration activation energy estimated using the 1/f noise measurements for different interconnects. The values of the electromigration activation energy for the Cu and Al interconnects determined from the 1/f noise measurements are 0.76−1.10 and 0.67−1.14 eV, respectively.21,24,37,59 Our data suggest that quasi-1D TaSe3 nanowires have similar $E_A$ values. Considering the 10x fold higher current carrying capacity and a possibility of ultimate downscaling of the quasi-1D TaSe3 van der Waals material, one can consider TaSe3 to be a promising material for the nanoscale interconnects.

One should note that the surface of the channel material can play an important role in defining the mechanism and level of 1/f noise. In the present study, a direct comparison between h-BN-capped and uncapped TaSe3 devices was not possible. The fabricated uncapped TaSe3 devices typically had non-Ohmic contacts and unstable I-V characteristics. The noise performance of such uncapped devices was affected by the current instability and non-Ohmic nature of the contacts. The effect of the surface on the noise characteristics in such a case cannot be accurately extracted, like it has been done for graphene, few-layer graphene, and MoS2 devices.28,29,60 However, because the thickness of TaSe3 channel in the tested devices is still relatively large, and the channel material is a metal, we believe that the 1/f noise response is primarily defined by the “volumetric” noise.

In conclusion, we investigated the low-frequency excess noise in quasi-1D nanowires of TaSe3-capped with quasi-2D h-BN layers. The TaSe3 nanowires have relatively low level of the 1/f noise in comparison with carbon nanotubes and other low-dimensional materials. The temperature-dependent measurements revealed that the electronic excess noise becomes the 1/f type with $\gamma > 1$ as temperature increases. Using the Dutta−Horn random fluctuation model of 1/f noise in metals and empirical model for metallic interconnects, we extracted the noise activation energy for quasi-1D TaSe3 nanowires. Our results indicate that quasi-1D van der Waals materials have potential for the ultimately downscaled local interconnect applications.

Methods. Material Preparation. TaSe3 was prepared from elemental tantalum (12.0 mmol, STREM, 99.98% purity) and selenium (35.9 mmol, STREM, 99.99% purity) with iodine (∼6.45 mg/cm3, J.T. Baker, 99.9% purity) as the transport agent. The tantalum/selenium mixture was ground and placed in a 17.78 × 1 cm fused quartz ampule (cleaned in concentrated nitric acid followed by annealing for 12 h at 900 °C). The charged reaction ampule was evacuated and backfilled with Ar 3× while submerged in an acetonitrile/dry ice bath. The flame-sealed ampule was placed in a three-zone, horizontal tube furnace heated at 20 °C min−1 to the final temperature gradient of 700 °C (hot zone)−680 °C (cool zone). The ampule was held at these temperatures for 2 weeks before cooling to room temperature. TaSe3 crystals were removed carefully from the quartz ampule, and residual I2 was removed by vacuum sublimation at 50 °C. The isolated yield of silver-black crystals was 90.8%. TaSe3 was chemically exfoliated by sonicating 6 mg of powdered TaSe3 crystals in 10 mL of ethanol for 4 h, resulting in a brownish-black suspension. This suspension was then centrifuged at 2600 rpm for 15 min, leaving well-dispersed TaSe3 wires 30−80 nm wide in the supernatant.

Table 1. $E_A$ and $E_P$ Extracted from 1/f Noise Measurements for Different Technologies

<table>
<thead>
<tr>
<th>Technology</th>
<th>$E_P$ eV (D−H model)</th>
<th>$E_A$ eV (empirical model)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu direct etch</td>
<td>1.10±4</td>
<td>0.79±7</td>
</tr>
<tr>
<td>Cu with TaNTa barrier</td>
<td>0.76−0.7824</td>
<td>0.7927</td>
</tr>
<tr>
<td>Al</td>
<td>0.69±1</td>
<td>0.80±12</td>
</tr>
<tr>
<td>Al−Si (1%)</td>
<td>0.80±11</td>
<td>0.89±21</td>
</tr>
<tr>
<td>Al−Cu (4%)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Al thin film</td>
<td>1.14±22</td>
<td></td>
</tr>
<tr>
<td>Al−Si (0.8%) passivated</td>
<td>0.74±39</td>
<td></td>
</tr>
<tr>
<td>Al−Si (1%)−Cu (0.5%) passivated</td>
<td>0.78±39</td>
<td></td>
</tr>
<tr>
<td>TaSe3</td>
<td>1.0 [this work]</td>
<td>0.88 [this work]</td>
</tr>
</tbody>
</table>

upon submission

AUTHOR INFORMATION

Corresponding Author
*E-mail balandin@ece.ucr.edu (A.A.B.).

ORCID
Guangxiong Liu: 0000-0001-8695-8671

Author Contributions
A.A.B. conceived the idea, coordinated the project, and contributed to experimental data analysis; T.T.S. supervised material synthesis and contributed to materials analysis; G.L. fabricated and tested devices, analyzed experimental data, and

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conducted part of the low-frequency noise measurements; M.A.B. synthesized TaSe$_3$ and conducted materials characterization; S.R. conducted noise measurements and analyzed experimental data; M.S. contributed to data analysis. All authors contributed to writing of the manuscript.

Notes
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