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High-frequency current oscillations in charge-density-wave 1T-TaS_2 devices: Revisiting the "narrow band noise" concept

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Adane K. Geremew,[1,a)] Sergey Rumyantsev,[1,2] Bishwajit Debnath,[3,a)] Roger K. Lake,[3] and Alexander A. Balandin[1,b)]

AFFILIATIONS

[1]Nano-Device Laboratory (NDL) and Phonon Optimized Engineered Materials (POEM) Center, Department of Electrical and Computer Engineering, University of California, Riverside, California 92521 USA
[2]Center for Terahertz Research and Applications (CENTERA), Institute of High-Pressure Physics, Polish Academy of Sciences, Warsaw 01-142, Poland
[3]Laboratory for Terascale and Terahertz Electronics (LATTE), Department of Electrical and Computer Engineering, University of California, Riverside, California 92521, USA

[a)]**Present address:** Intel Corporation, Hillsboro, Oregon, USA.
[b)]**Author to whom correspondence should be addressed**: balandin@ece.ucr.edu. **URL:** http://balandingroup.ucr.edu/

ABSTRACT

We report on the current oscillations in quasi-2D 1T-TaS_2 charge-density-wave two-dimensional devices. The MHz-frequency range of the oscillations and the *linear* dependence of the frequency of the oscillations on the current closely resemble the narrow band noise, which was often observed in the *classical* bulk quasi-1D trichalcogenide charge-density-wave materials. In bulk quasi-1D materials, the narrow band noise was interpreted as *direct* evidence of charge-density-wave sliding. Despite the similarities, we argue that the nature of the MHz oscillations in 1T-TaS_2 is different from the narrow band noise. Analysis of the biasing conditions and current indicates that the observed oscillations are related to the current instabilities due to the voltage-induced transition from the nearly commensurate to incommensurate charge-density-wave phase.

It is well known since the mid-20th century that some materials, usually metals, with quasi-one-dimensional (1D) crystal structure can form the charge-density-wave (CDW) state.[1] Examples of such materials, many of them transition metal trichalcogenides (TMTs), include NbS_3 and TaS_3. A large number of interesting CDW-related effects have been observed experimentally and studied theoretically.[1–9] They include nonlinear electrical conductivity, phase transitions induced by temperature and electric field, and de-pinning and sliding of CDWs under applied electric bias. One of the most interesting phenomena observed for quasi-1D bulk CDW materials was the narrow band noise.[2–9] It was found that at a critical bias voltage V_T corresponding to a critical electric field, the noise spectrum in the low MHz frequency range reveals one or several peaks. The effect was associated with the de-pinning and sliding of the CDWs. The peaks in the noise spectra were interpreted as an indication of the periodic current oscillations due to the collective current of the sliding CDW. The term "noise" in the present context of periodic oscillations is not quite correct.[1] However, it has been used extensively in literature, and we mention it here for a historic prospect.

The studies of the current oscillations, i.e., narrow band noise, in bulk 1D CDW materials observed a linear relationship between the frequency, f, and the collective, i.e., excess, current, $I_{CDW} = I - I_N$, attributed to the CDW sliding, where I is the total measured current and I_N is the conventional current due to free electrons, extracted from Ohm's law. The linear relationship was explained by the assumption that f is proportional to the CDW drift velocity, v_D, so that $f = v_D/\Lambda$, where Λ is the characteristic distance.[2,3] Since $I_{CDW} = nef\Lambda A$, where n is the charge carrier density, e is the charge of an electron, and A is the cross-sectional area of the sample, one can write for the frequency $f = (1/ne\Lambda A) \times I_{CDW}$.[1] Other models, with more material specificity, resulted in a similar relationship between the frequency and current. The observation of the narrow band noise was interpreted as the direct evidence of the CDW current, which arises from the sliding of CDWs. The narrow band noise was also related to a special new type of instability and negative differential dielectric constant.

Recent years witnessed a rebirth of the CDW field.[10–20] It has been associated, from one side, with the interest in layered quasi-two-dimensional (2D) van der Waals materials and, from another side, with the realization that some of these materials reveal CDW effects at room temperature (RT) and above.[16] Examples of these materials, many of them transition metal dichalcogenides (TMDs), include the 1T polymorphs of TaS_2 and $TaSe_2$. The most interesting 2D layered TMD with CDW properties is the 1T polymorph of TaS_2. It undergoes the transition from a normal metallic phase to an incommensurate CDW (IC-CDW) phase at 545 K, then to a nearly commensurate CDW (NC-CDW) phase at 350 K, and, finally, to a commensurate CDW (C-CDW) phase at ∼200 K.[10–16] The C-CDW phase consists of a $\sqrt{13} \times \sqrt{13}$-reconstruction within the basal plane that forms a star-like pattern, in which each star contains 13 Ta atoms. The Fermi surface, composed of 1D-electron per star, is unstable. The lattice reconstruction is accompanied by a Mott–Hubbard transition that gaps the Fermi surface and increases the resistance.

As the temperature increases, the C-CDW phase breaks up into the NC-CDW phase, which consists of the C-CDW domains. The C-CDW to NC-CDW transition is revealed as an abrupt change in the resistance with a large hysteresis window in the resistance profile at ∼200 K. As the temperature is increased to ∼350 K, the NC-CDW phase melts into the I-CDW phase. This transition is accompanied by a smaller hysteresis window in the resistivity. It has been conformed in numerous studies that 1T-TaS_2-quasi-2D CDW quantum material—reveals numerous intriguing properties, including multiple phase transitions, accompanied by resistance change and hysteresis, dependence of the phase transition points on the number of atomic planes, and "hidden" phases at low temperature.[10–16] Intercalation, doping, pressure, optical excitation, and electric source–drain bias can modify the transitions to different CDW phases in 1T-TaS_2. The demonstrated RT operation of CDW oscillators,[16] possible transistor-less circuits on the basis of CDW devices,[17,18] and intriguing radiation hardness against x-ray and protons[19,20] moved 2D CDW research into the domain of applied physics and practical applications.

Despite the strong interest in 1T-TaS_2 and other 2D materials, narrow band noise has never been observed in such materials. The *current fluctuations*, i.e., narrow band noise, which was interpreted as a direct evidence of CDW de-pinning and sliding in "classical" bulk quasi-1D CDW materials,[1–9] is missing in the "new generation" quasi-2D CDW materials. In our previous studies of 1T-TaS_2, we focused on the low-frequency noise, termed the "broad band noise" in the CDW community,[1] conducting measurements from 1 Hz to ∼100 kHz.[21–23] We observed peaks in the noise spectral density as a function of bias at the frequencies below 100 kHz, which were related to the CDW phase transitions, de-pinning, and possible "hidden phases."[21–23] However, no current fluctuations, i.e., narrow band noise in the MHz or other spectral range, have ever been reported for 1T-TaS_2 or other quasi-2D CDW materials. In this letter, we report an observation of current fluctuations in 1T-TaS_2 in the MHz frequency range, which have a striking similarity to the narrow band noise in the quasi-1D CDW materials such as NbS_3. In contrast to the quasi-1D materials, we find that the noise peaks in the quasi-2D CDW test structures can be explained by the hysteresis at the IC-CDW–NC-CDW phase transitions rather than by sliding of CDWs. We offer a possible explanation for the absence of the *true* narrow band noise in quasi-2D CDW materials.

Thin films of crystalline 1T-TaS_2 were mechanically exfoliated and transferred to SiO_2/Si substrates. The thickness of SiO_2 layer was always 300 nm. The metal Ti/Au contacts were evaporated through a shadow mask to avoid chemical contamination and oxidation during the fabrication process. The shadow masks were fabricated using double-side polished Si wafers with 3 μm thermally grown SiO_2 (Ultrasil Corp.; 500-μm thickness; P-type; ⟨100⟩). For the devices used in the study, the 1T-TaS_2 thickness was in the range from 10 nm to 50 nm, the channel length was about 2 μm, and the width ranged from 10 μm to 15 μm. Details of fabrication of 1T-TaS_2 two-terminal devices, i.e., test structures, have been reported by us elsewhere.[21,22] The current–voltage (I–V) characteristics and resistivities were measured with a semiconductor analyzer (Agilent B1500). The noise measurements were conducted under DC bias voltages, V_{SD}, in the frequency range from 50 kHz to 2 MHz. The voltage fluctuations from the 1 kΩ load resistor were amplified by a low noise amplifier (Stanford Research SR 560) and analyzed by a spectrum analyzer (Brüel and Kjær). The value of the load resistance was selected to be close to that of the device under test as normally done in the noise measurements. Since the nominal bandwidth of the amplifier (SR 560) is 1 MHz, the gain at higher frequencies was measured separately as a function of frequency, and the actual gain at given frequency was used to plot the spectra. More information on our noise testing protocols can be found in prior publications.[21–26]

Figure 1 shows the dependence of noise spectral density, S_I, on the current through the device channel at frequency $f = 760$ kHz. To ensure the reproducibility, the data are shown for two test structures. The general trend is $S_I \propto I^2$, as one typically observes in the low-frequency noise measurements for various materials.[27,28] However, one can notice some deviation from the I^2 scaling, i.e., steeper increase in S_I with the current. At low current levels, $I < 8 \times 10^{-4}$ A, the noise

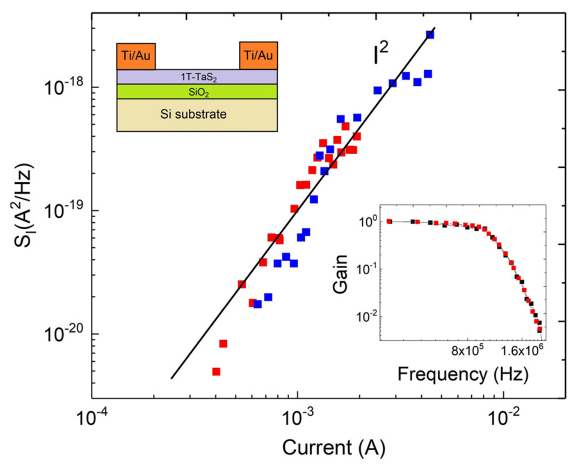

FIG. 1. Noise power spectral density, S_I, as a function of the current through 1T-TaS_2 device channel measured at frequency $f = 760$ kHz. The red and blue data points correspond to two tested devices. The upper inset shows a schematic of the two-terminal CDW devices; the lower inset shows the gain, normalized to the gain at $f = 30$ kHz, as a function of frequency.

spectral density, S_I, had the form of the $1/f$ noise, i.e., $S_I(f) \propto 1/f$. As the current increased, the spectral density became flatter, possibly indicating the increasing contribution of the generation-recombination type noise, which has a Lorentzian spectrum. This spectrum flattening is likely responsible for S_I deviation from I^2 scaling. The insets to Fig. 1 show the device schematic and the normalized dependence of the gain on frequency.

The measurements of the noise spectral density, S_I, as the function of frequency, f, at higher currents, $I > 2$ mA, demonstrated the peak, closely resembling the narrow band noise observed in bulk quasi-1D CDW materials.[1–9] Figure 2 shows the noise spectra close to the peak for a representative device at different currents. The spectra were obtained by normalizing the measured spectra by the amplitude–frequency characteristic shown in the inset to Fig. 1. As one can see, the frequency of the peak increases with increasing current similar to the current dependence of the narrow band noise, reported in numerous publications for bulk samples of quasi-1D CDW materials.[1–9] Each peak in Fig. 2 corresponds to the periodic current oscillations with the fundamental frequency f_{max}.

The dependence of the current oscillation frequency, f_{max}, on the current is presented in Fig. 3. The extracted current dependence of the oscillation frequency is close to linear. As we discussed above, the frequency of the narrow band noise depends linearly on the current according to different models, i.e., $f = (1/ne\Lambda A) \times I_{CDW}$.[1–9] In this sense, the observed noise peaks in 1T-TaS_2 devices in the MHz frequency range closely resemble the narrow band noise in the bulk quasi-1D CDW materials such as NbS_3. However, there is one substantial difference with the classical narrow band noise experiments. The current density in our experiments, at which the peaks are observed, is on the order of 10^6 A/cm^2, which is many orders of magnitude larger than in narrow band noise experiments.[1] For this reason, the mechanism of the current oscillations in quasi-2D 1T-TaS_2 might be different.

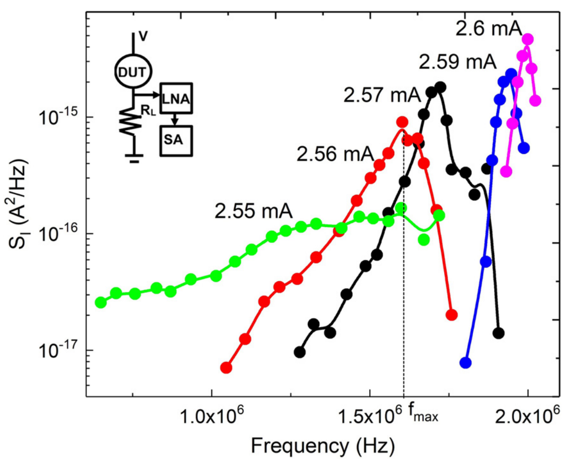

FIG. 2. Noise power spectral density as a function of frequency for several value of the current through the device channel. The spectra were obtained by normalizing the measured spectra by the amplitude-frequency characteristic shown in the inset in Fig. 1. The peak shifts to the higher frequency f_{max} with the increasing current, closely resembling the narrow band noise observed for bulk samples of quasi-1D CDW materials. The inset shows the schematic of the measurement.

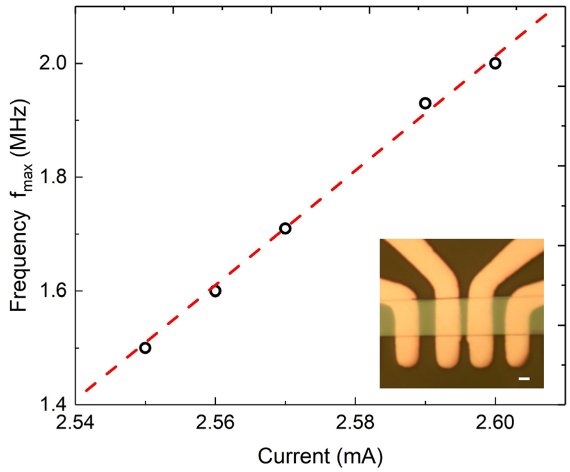

FIG. 3. Frequency, f_{max}, of the noise peaks as a function of the current through 1T-TaS_2 device channel. The narrow band noise in bulk quasi-1D CDW materials reveled a linear dependence of the oscillation frequency on the current. The inset shows a microscopy image of a representative 1T-TaS_2 device structure with several metal contacts.

We hypothesized that the current oscillations of a particular frequency in the MHz regime can be not the signatures of the CDW sliding but IC-CDW–NC-CDW phase transition similar to the one used by the present authors for demonstration of a CDW voltage-controlled oscillator.[16] Figure 4 shows the I–V characteristic of the same 1T-TaS_2 device used to demonstrate the noise peaks. The dashed line represents the load of the 1 kΩ resistor, and for one of the total applied voltages used in the experiments. The hysteresis loop with the on-set at 0.8 V is the phase transition from NC-CDW to IC-CDW induced by the applied electric bias to the device channel. This type of phase transition is well known, and it has been reported in various

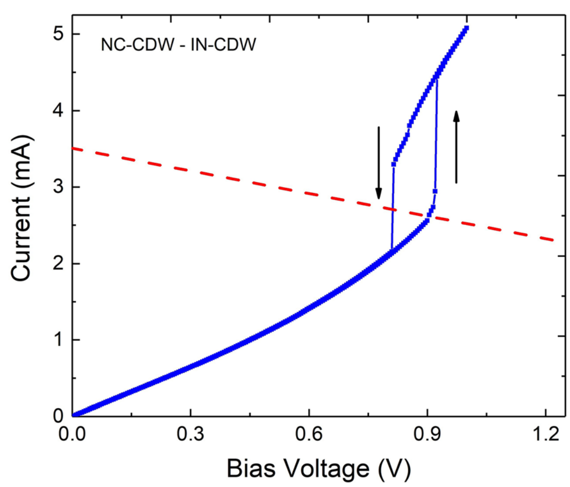

FIG. 4. Current–voltage characteristics of tested 1T-TaS_2 device (the same as in Fig. 2). The hysteresis loop at the bias voltage V = 0.9 V corresponds to the transition from the NC-CDW phase to the IC-CDW phase induced the applied electric field. The arrows indicate the forward and reverse cycle currents.

independent studies, including by means of the noise spectroscopy.[12,16,21] At this bias, the circuit has two working points at different voltages and currents. The state with the smaller voltage on the sample corresponds to the higher current. Although the details of phase transition dynamics are not well known,[12] we speculate that the observed current oscillations result from the instability at the voltages close the phase transition hysteresis loop. This mechanism is different from the narrow band noise observed in quasi-1D bulk CDW materials, where it was attributed to the CDW sliding.[1]

The scaling of the frequency of the oscillations with the current based on the NC-CDW–IC-CDW phase transition can be affected by the parasitic elements of the device test structure and measurement setup. A circuit model of the 1T-TaS_2 sample, load resistance, and parasitic capacitance is shown in the inset of Fig. 5.[16] The 1T-TaS_2 is modeled as a state-dependent resistance R_D with values $R_m = 232.3\ \Omega$ ("metallic") and $R_i = 946.5\ \Omega$ ("insulating"). The 1T-TaS_2 sample is biased with an external load resistance, $R_s = 1\,\mathrm{k}\ \Omega$, and a parasitic capacitance, $C = 1.2$ nF.[16] The CDW transition creates a bistability that causes the RC circuit to oscillate. The charging and discharging equations of the circuit are

$$C\frac{dV_o}{dt} = \begin{cases} (V_{DD} - V_o)/R_S - V_o/R_i : & \text{Charging} \\ (V_{DD} - V_o)/R_S - V_o/R_m : & \text{Discharging}, \end{cases} \qquad (1)$$

where V_{DD} is the supply voltage and V_o is the output voltage as shown in the inset of Fig. 5. The circuit oscillates between V_L and V_H defined by the low and high voltages of the bistable region shown in Fig. 4. The state-dependent resistance R_D of the 1T-TaS_2 for voltages outside of the bistable region is

$$R_D = \begin{cases} R_i & (V_o < V_L) \\ R_m & (V_o > V_H), \end{cases}$$

and inside the bistable region it is

$$R_D = \begin{cases} R_i & (dV_o/dt > 0) \\ R_m & (dV_o/dt < 0). \end{cases}$$

We solved Eq. (1) numerically for $V_o(t)$ by discretizing the time derivative, using a time step of 2.43 ns, and an initial condition $V_o(t = 0) = 0$. The resulting time-dependent output voltage is Fourier transformed, and the fundamental frequency peak is plotted in Fig. 5. An example spectrum is shown in the upper left inset. At low currents, such as those used in the experiment, the frequency of the fundamental peak increases linearly with the average DC.

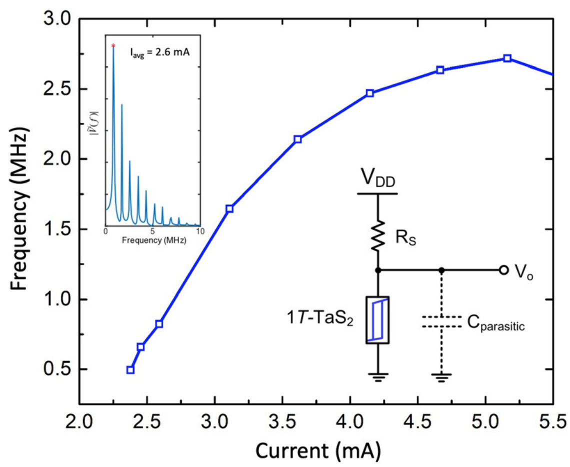

FIG. 5. Calculated peak frequency of the voltage oscillations vs the DC from the source. The circuit is shown in the lower right inset, and an example frequency spectrum is shown in the upper left inset. The maximum frequency marked with the red star is plotted in the main plot.

The intriguing question which remains is why nobody has observed the true narrow band noise in quasi-2D CDW TMD materials such as 1T-TaS_2. One possibility can be that in devices operating in the NC-CDW phase, the collective CDW current conduction is always overshadowed by the conventional individual electron conduction because the full gapping in 1T-TaS_2 is only expected at low temperature, below the C-CDW transition. There are different opinions on the importance of IC-CDW phase for observation of depinning and sliding. In our previous studies of the low-frequency noise in 1T-TaS_2, we observed the signatures of depinning and sliding in NC-CDW phase,[21,22] before the onset of the NC-CDW to IC-CDW transition. In the present study, we have not observed narrow band noise at the bias above the hysteresis window of the NC-CDW–IC-CDW transition. Another possibility can be that the characteristic distance, Λ, in the equation for the current oscillation, $f = (1/ne\Lambda A) \times I_{CDW}$, which can be associated with the CDW coherence length[2,3] is shorter, and much broader distributed, in 2D layered material than in 1D layered materials. The third, most drastic possibility, which would require systematic re-investigation of CDW effects in bulk quasi-1D TMT materials, is that the presence of hysteresis may have been overlooked at least in some earlier studies. The CDW de-pinning is always accompanied by the nonlinear conduction and, in some cases, by hysteresis effects. This hysteresis close to the voltage of the CDW de-pinning can lead to the oscillations, similar to the situation observed in the present study. The narrow band noise was typically observed using noise methods and revealed itself as oscillations above the background noise. Similar oscillations can be observed just due to the hysteresis at the offset to de-pinning and not by the sliding itself. Since the amplitude of the oscillations is small, the small hysteresis, which causes these oscillations, could have been overlooked in the *I–V* characteristics measurements. At least a few studies of bulk quasi-1D CDW materials discuss the narrow hysteresis at the critical depinning field.[29,30]

In summary, intrigued by the absence of the reports of the narrow band noise in quasi-2D CDW materials, we undertook the investigation of the noise response of the 1T-TaS_2 thin-film test structure to the MHz frequency range. At certain values of the current through the two-terminal devices, we found the current oscillations, with the frequency proportional to the value of the current, which resemble closely the classical narrow band noise bulk quasi-1D trichalcogenide CDW materials. However, despite the similarities, we argued that the nature of the MHz oscillations in 1T-TaS_2 is different, and it is related to the current instabilities at the NC-CDW to IC-CDW transition rather than to the CDW sliding. The obtained results are important for better understanding of electron transport and phase transitions in quasi-2D CDW materials and for their proposed applications in the radiation hard electronics.

AUTHOR'S CONTRIBUTIONS

A.A.B. and S.R. conceived the idea. A.B. coordinated the project, and led the data analysis and manuscript preparation; A.K.G. fabricated the devices, conducted electrical measurements, and assisted with the noise measurements; S.R. conducted the noise measurements and data analysis; B.D. performed the numerical circuit simulations; R.L. contributed to the circuit and data analysis. All authors contributed to the manuscript preparation.

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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REFERENCES

[1]For a review, see G. Grüner, Rev. Mod. Phys. **60**, 1129 (1988); and references therein.
[2]R. M. Fleming and C. C. Grimes, Phys. Rev. Lett. **42**, 1423 (1979).
[3]G. Grüner, A. Zettl, W. G. Clark, and A. H. Thompson, Phys. Rev. B **23**, 6813 (1981).
[4]M. Oda and M. Ido, Solid State Commun. **50**, 879 (1984).
[5]K. Seeger, Solid State Commun. **53**, 219 (1985).
[6]B. Horovitz, A. R. Bishop, and P. S. Lomdahl, Physica B+C **143**, 135 (1986).
[7]T. M. Tritt, D. J. Gillespie, and A. C. Ehrlich, Phys. Rev. B **41**, 7948 (1990).
[8]F. Y. Nad and P. Monceau, Phys. Rev. B **46**, 7413 (1992).
[9]D. DiCarlo, J. McCarten, T. L. Adelman, M. P. Maher, and R. E. Thorne, Phys. Rev. B **49**, 14722 (1994).
[10]F. Zwick, H. Berger, I. Vobornik, G. Margaritondo, L. Forró, C. Beeli, M. Onellion, G. Panaccione, A. Taleb-Ibrahimi, and M. Grioni, Phys. Rev. Lett. **81**, 1058 (1998).
[11]T. Pillo, J. Hayoz, H. Berger, R. Fasel, L. Schlapbach, and P. Aebi, Phys. Rev. B **62**, 4277 (2000).
[12]B. Sipos, A. F. Kusmartseva, A. Akrap, H. Berger, L. Forró, and E. Tutiš, Nat. Mater. **7**, 960 (2008).
[13]M. J. Hollander, Y. Liu, W. J. Lu, L. J. Li, Y. P. Sun, J. A. Robinson, and S. Datta, Nano Lett. **15**, 1861 (2015).
[14]L. Stojchevska, I. Vaskivskyi, T. Mertelj, P. Kusar, D. Svetin, S. Brazovskii, and D. Mihailovic, Science **344**, 177 (2014).
[15]L. Ma, C. Ye, Y. Yu, X. F. Lu, X. Niu, S. Kim, D. Feng, D. Tománek, Y.-W. Son, X. H. Chen, and Y. Zhang, Nat. Commun. **7**, 10956 (2016).
[16]G. Liu, B. Debnath, T. R. Pope, T. T. Salguero, R. K. Lake, and A. A. Balandin, Nat. Nanotechnol. **11**, 845 (2016).
[17]A. Khitun, G. Liu, and A. A. Balandin, IEEE Trans. Nanotechnol. **16**, 860 (2017).
[18]A. G. Khitun, A. K. Geremew, and A. A. Balandin, IEEE Electron Device Lett. **39**, 1449 (2018).
[19]G. Liu, E. X. Zhang, C. D. Liang, M. A. Bloodgood, T. T. Salguero, D. M. Fleetwood, and A. A. Balandin, IEEE Electron Device Lett. **38**, 1724 (2017).
[20]A. K. Geremew, F. Kargar, E. X. Zhang, S. E. Zhao, E. Aytan, M. A. Bloodgood, T. T. Salguero, S. Rumyantsev, A. Fedoseyev, D. M. Fleetwood, and A. A. Balandin, Nanoscale **11**, 8380 (2019).
[21]G. Liu, S. Rumyantsev, M. A. Bloodgood, T. T. Salguero, and A. A. Balandin, Nano Lett. **18**, 3630 (2018).
[22]A. K. Geremew, S. Rumyantsev, F. Kargar, B. Debnath, A. Nosek, M. A. Bloodgood, M. Bockrath, T. T. Salguero, R. K. Lake, and A. A. Balandin, ACS Nano **13**, 7231 (2019).
[23]R. Salgado, A. Mohammadzadeh, F. Kargar, A. Geremew, C.-Y. Huang, M. A. Bloodgood, S. Rumyantsev, T. T. Salguero, and A. A. Balandin, Appl. Phys. Express **12**, 037001 (2019).
[24]M. A. Stolyarov, G. Liu, S. L. Rumyantsev, M. Shur, and A. A. Balandin, Appl. Phys. Lett. **107**, 023106 (2015).
[25]J. Renteria, R. Samnakay, S. L. Rumyantsev, C. Jiang, P. Goli, M. S. Shur, and A. A. Balandin, Appl. Phys. Lett. **104**, 153104 (2014).
[26]G. Liu, S. Rumyantsev, M. S. Shur, and A. A. Balandin, Appl. Phys. Lett. **102**, 093111 (2013).
[27]P. Dutta and P. M. Horn, Rev. Mod. Phys. **53**, 497 (1981).
[28]V. Mitin, L. Reggiani, and L. Varani, "Generation-recombination noise in semiconductors," in *Noise and Fluctuations Control in Electronic Devices*, edited by A. A. Balandin (American Scientific Publishing, Los Angeles, 2003).
[29]A. Zettl and G. Gruner, Phys. Rev. B **26**, 2298 (1982).
[30]M. E. Itkis, F. Y. Nad, and P. Monceau, Synth. Met. **41-43**, 4037 (1991).