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Final Technical Report

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Project Title: Physical Mechanisms and Electric-Bias Control of Phase Transitions in Quasi-2D Charge-Density-Wave Quantum Materials

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08/15/2020 - 08/14/2023
08/15/2020 - 05/15/2024 (with no cost extension)

Research Goals and Scope

This DOE project aims to understand the physical mechanisms and develop methods for electric-bias control of phase transitions in quasi-two-dimensional (2D) charge-density-wave (CDW) quantum materials. The investigation mostly focuses on thin films of transition metal dichalcogenides (TMD), *e.g.* TaS_2, which reveal phase transitions at room temperature (RT) and above. The specific objectives of the project include the development of innovative approaches for investigation and controlling CDW phases with external stimuli; understanding the physical mechanisms behind the phase transitions in quasi-2D van der Waals materials; investigating the "hidden phases" at temperatures below the transition to commensurate CDW phase; and separating the electric field CDW switching from Joule heating induced switching. The physics of CDW phases in quasi-2D materials of the TMD group is substantially different from and much less understood than their bulk counterparts with the quasi-one-dimensional (1D) crystal structure. In addition to the above RT phase transitions, 1T-TaS_2 reveals such intriguing properties as multiple step-like resistance changes hysteresis. Reports on the de-pinning and sliding of CDWs in 2D material systems are scarce. The electric field's and local heating's role in inducing the transitions in 2D CDW is not completely clear. The use of innovative characterization techniques such as low-frequency noise spectroscopy, Brillouin and Raman light scattering spectroscopy, and ultra-fast current pulses are utilized to investigate and understand these phenomena.

Recent Results

In the reported period (no-cost extension), we investigated the temperature dependence of the current fluctuations in thin films of the quasi-two-dimensional 1T-TaS_2 van der Waals material. The current fluctuations, determined from the derivative current-voltage characteristics of two-terminal 1T-TaS_2 test structures, appear prominently at the electric fields that correspond to the transitions between various CDW condensate phases and at the onset of the depinning of the charge density wave domains. The depinning threshold field monotonically increases with decreasing temperature within the nearly commensurate CDW phase. The threshold field value increases with the decreasing 1T-TaS_2 film thickness, revealing the surface pinning of the CDWs. Our analysis suggests that the domain depinning is pronounced in the nearly commensurate phase. It is induced by the electric field but facilitated by local heating. The measured trends for the threshold field of the domain depinning are important for understanding the physics of charge density waves in quasi-two-dimensional crystals [1]. While the majority of the studies were conducted with test structures based on individual films of 1T-TaS_2, we have also experimented with the composites that had 1T-TaS_2 flakes as fillers. This allowed us to verify that the CDW phase transitions observed in bulk crystals and individual films of 1T-TaS_2 can persist in the composites. This aspect of work allowed us to determine that the phase transition between the nearly commensurate and incommensurate CDW phases is robust against disorder [2].

Highlights of the Main Results

In this project, we investigated in detail the depinning of nearly commensurate CDW in 1T-TaS_2 thin films at room temperature. A combination of the differential current-voltage measurements with the low-frequency noise spectroscopy provided a means for detecting the depinning threshold field in quasi-2D materials. We established that the depinning process in 1T-TaS_2 is not accompanied by an observable abrupt increase in electric current – in contrast to depinning in the conventional CDW materials with quasi-1D crystal structure [3]. We explained it by the fact that the current density from the CDWs in the 1T-TaS_2 devices is orders of magnitude smaller than the current density of the free carriers available in the dis-commensuration network surrounding the commensurate CDW islands. The depinning fields in 1T-TaS_2 thin-film devices are several orders of magnitude larger than those in quasi-1D van der Waals materials. We have also investigated the room-temperature switching of 1T-TaS_2 thin-film CDW test structures, using nanosecond-duration electrical pulsing to construct their time-resolved current-voltage characteristics [4]. The switching action was based upon the nearly commensurate to incommensurate CDW phase transition in this material, which has a characteristic temperature of 350 K at thermal equilibrium. For sufficiently short pulses, with rise times in the nanosecond range, the self-heating of the devices was suppressed, and their current-voltage characteristics were weakly nonlinear and free of hysteresis. This changed as the pulse duration was increased to 200 ns, where the current developed pronounced hysteresis that evolved nonmonotonically with the pulse duration. By combining the results of our experiments with a numerical analysis of transient heat diffusion in these devices, we revealed the thermal origins of their switching.

We achieved the electrical gating of the CDW phases and currents in h-BN capped three-terminal 1T-TaS_2 heterostructures [5]. Electrical gating is important for answering a fundamental science question: "Can one achieve an electrical switching of CDW phase *via* the pure field effect, without any local Joule heating involved?" The main difficulty of electrical gating of the CDW phases and currents in 2D van der Waals materials was associated with the fact that different CDW phases in 2D materials still have a rather high concentration of charge carriers. Below 550 K, 1T-TaS_2 is in the metallic-like incommensurate CDW (IC-CDW) phase. The nearly commensurate phase (NC-CDW) appears below 350 K and persists approximately until 180 K. Below this temperature, 1T-TaS_2 enters the commensurate CDW (C-CDW) phase. The high concentration of charge carriers results in a small relative change of the carrier concentration due to the large number of carriers and strong screening of the gate potential. Reducing the thickness of 1T-TaS_2 thin film for a stronger gating effect is not necessarily a viable approach because at small thicknesses the CDW phases can be locked and some phase transitions disappear. We have successfully demonstrated that the application of a gate bias in the h-BN/1T-TaS_2 structure can shift the source-drain current-voltage hysteresis associated with the transition between the nearly commensurate and incommensurate CDW condensate phases. The evolution of the hysteresis and the presence of abrupt spikes in the current while sweeping the gate voltage indicated that the effect was electrical rather than self-heating. We attributed the gating to an electric-field effect on the commensurate CDW domains in the atomic planes near the gate dielectric. The transition between the nearly commensurate and incommensurate CDW phases can be induced by both the source-drain current and the electrostatic gate.

In the course of this work, we also verified the CDW phases can be preserved and switched in the solution-processed 1T-TaS_2 thin films [6]. The inks were prepared by liquid-phase exfoliation of CVT-grown 1T-TaS_2 crystals to produce fillers with nm-scale thickness and micrometer-scale lateral dimensions. The temperature-dependent electrical and current fluctuation measurements of printed thin films demonstrated that the CDW properties of 1T-TaS_2 are preserved after processing. The functionality of the thin-film devices was defined by the nearly-commensurate to commensurate CDW phase transitions in the individual exfoliated 1T-TaS_2 fillers rather than by electron-hopping transport between them. The low-frequency noise spectroscopy was used for monitoring CDW phase transitions.

Highlights of the Conference Talks

The results of the project were disseminated *via* journal publications and conference presentations. The highlights of the talks, with the published results, relevant to this project include.

- A.A. Balandin, invited talk, "Charge-density-wave phase transitions in quasi-2D 1T-TaS_2 devices," Low-Dimensional Materials and Devices Conference, SPIE Optics + Photonics, San Diego, USA, 2022
- J.O. Brown, M. Taheri, N. R. Sesing, T. T. Salguero, F. Kargar, and A. A. Balandin, "Charge-density-wave phase transitions in quasi-2D 1T-TaS_2/h-BN heterostructure devices," SPIE Nanoscience + Engineering, San Diego, California, USA, 2022.
- M. Taheri, J. Brown, F. Kargar, and A. A. Balandin, "Electrical gating of the charge-density-waves in two-dimensional 1T-TaS_2 devices," in Workshop on Innovative Nanoscale Devices and Systems (WINDS), December 2022, Lihue, Hawaii, USA.
- J.O. Brown, M. Taheri, F. Kargar, R. Salgado, T. Geremew, S. Rumyantsev, R. K. Lake, and A. A. Balandin, "Temperature dependence of the charge-density-wave depinning in 1T-TaS_2 devices," APS March Meeting, Las Vegas, Nevada, USA, 2023
- M. Taheri, J. Brown, A. Rehman, N. R. Sesing, F. Kargar, T. T. Salguero, S. Rumyantsev, A. A. Balandin, "Electrical gating of the quantum condensate phases in two-dimensional charge-density-wave materials", MRS Meeting, Spring 2023, San Francisco, USA
- A.A. Balandin, invited talk, "2D charge-density-wave materials and devices," 7th Annual U.S. Government Workshop on 2D Materials, National Institute of Standards and Technology, USA, 2024 (virtual)

Summary

The goals of this fundamental science project, aimed at understanding the physical mechanisms and developing methods for electric-bias control of phase transitions in quasi-2D CDW materials, have been achieved. We focused on 1T-TaS_2, one of the most interesting materials of this type, and demonstrated electrical gating of the I-V characteristics and hysteresis in this material. The demonstration of electrical gating of CDW phases in quasi-2D material was performed at RT. We have conducted experiments to separate the electric-field CDW switching from Joule heating-induced switching. This was an important development for 2D CDW materials. The project has led to a better understanding of the physical mechanisms behind the phase transitions and CDW depinning in quasi-2D van der Waals materials. We established that the CDW domain depinning in 1T-TaS_2 does not lead to a strong increase in the collective current and accompanying narrow-band noise. We developed a technique that utilized the low-frequency noise measurements in such materials for monitoring the CDW phase transitions. In the experiments where 1T-TaS_2 flakes were used in polymeric matrices, we verified the robustness of the phase transitions between the nearly commensurate and incommensurate CDW phases.

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