Doping-Tunable Charge Ordering in Semiconducting Single-Layer Cr₂Se₃

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Abstract

The charge density wave (CDW), characterized spatially modulated by electronic charge density coupled with structural distortions, offers a valuable framework for exploring electron-electron interactions, electron-phonon coupling, and quantum phase transitions. Although extensively studied in group-IV and group-V transition metal chalcogenides (TMCs), CDW phenomena remain underexplored in other groups. Additionally, while the tunability of CDW has been investigated on macroscopic and mesoscopic scales, ^[1,2] atomic-scale visualization is seldom reported. Here, we present the real-space observation of a doping-tunable charge ordering phase in semiconducting single-layer Cr₂Se₃, a group-VI TMC. Using scanning tunnelling microscopy and spectroscopy (STM/STS), we observed charge ordering manifesting as granular patterns at low temperatures, featuring contrast inversion ^[3] in STM images with opposite bias voltages. The presence of a small bandgap at the Fermi level, with band edges modulated by the granular pattern, substantiates the charge ordering origin. Temperature-dependent STM and nc-AFM measurements revealed a phase transition and lattice distortion linked to charge ordering. The semiconducting nature of Cr₂Se₃ allows modulation of charge ordering through doping: hole doping suppresses charge orderina,

whereas electron doping modifies the pattern, yielding a periodic $3\sqrt{3} \times 3\sqrt{3}$ CDW phase. The observation of tunable charge ordering in a group-VI TMC enhances our understanding of the interplay between charge doping and charge ordering tunability in 2D materials.

References

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- [3] Spera, M., et al. Phys. Rev. Lett., 125 (2020), 267603.



Figure 1: (a) STM topographic image of singlelayer (SL) Cr2Se3/graphite. (b) Temperaturedependent dI/dV spectra of SL Cr₂Se₃. (c, d) Atomic resolution STM image of SL Cr₂Se₃ acquired at (c) 4.3 K and (d) 77 K, showing temperature-dependent features.

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