

Supplementary information: Giant tunability of superlattice excitations in chiral $\text{Cr}_{1/3}\text{TaS}_2$

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CONTENTS

S1. Loading the diamond anvil cell and assuring quasi-hydrostatic conditions	S-3
S2. Raman scattering response in the diamond anvil cell compared with measurements outside the cell	S-3
S3. Pristine vs. pressure-cycled materials	S-4
S4. Tunability of the metal monolayer excitation	S-4
References	S-5

S1. LOADING THE DIAMOND ANVIL CELL AND ASSURING QUASI-HYDROSTATIC CONDITIONS

Figure S1 displays a photograph of $\text{Cr}_{1/3}\text{TaS}_2$ inside of a diamond anvil cell with KBr as the pressure medium, an annealed ruby ball, and a gasket. The ruby allows for a direct measurement of pressure during the experiment because the $R1$ fluorescence shifts linearly under pressure. The shape of the ruby fluorescence also reveals whether the conditions inside the cell are hydrostatic (or not). If non-hydrostatic conditions arise, the R_1 and R_2 peaks will develop an asymmetric shape, or the peaks will merge together. As shown in Fig. S2, quasi-hydrostatic conditions are present across the full pressure range of our study.

Figure S1. *Photograph of the $\text{Cr}_{1/3}\text{TaS}_2$ crystal inside of the diamond anvil cell taken through a microscope at room temperature. In addition to the crystal, we can see an outline of the annealed ruby balls and the KBr pressure medium along with a 200 μm drilled gasket hole - all sitting on top of a 500 μm ultra-low fluorescence diamond culet. The green 532 nm laser is defocussed to avoid damaging the crystal.*

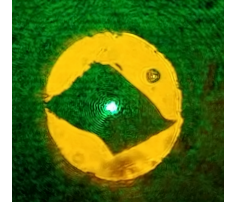
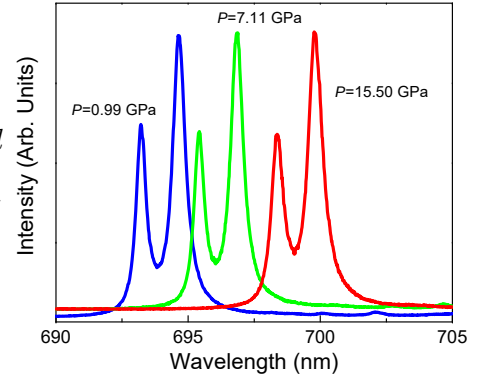


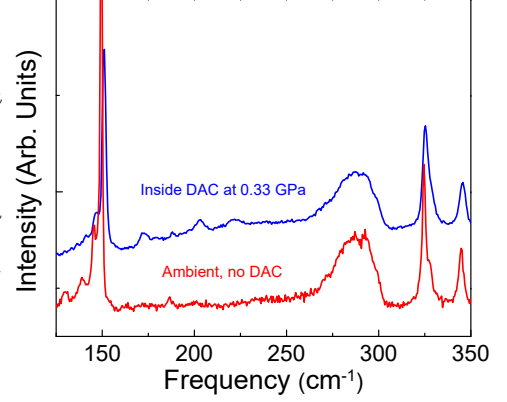
Figure S2. *Fluorescence of the annealed ruby ball at selected pressures at 300 K. The results are consistent with quasi-hydrostatic pressure conditions.*



S2. RAMAN SCATTERING RESPONSE IN THE DIAMOND ANVIL CELL COMPARED WITH MEASUREMENTS OUTSIDE THE CELL

Small out-of-plane features sometimes appear in the Raman scattering spectrum when $\text{Cr}_{1/3}\text{TaS}_2$ is measured in the diamond anvil cell [Fig. S3]. This is likely because the crystal is not perfectly flat inside the cell (as it is when the sample is measured independently).

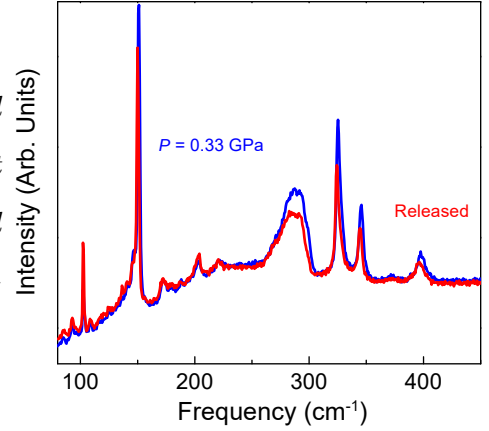
Figure S3. Raman scattering response of $\text{Cr}_{1/3}\text{TaS}_2$ at room temperature inside the diamond anvil cell ($P = 0.33 \text{ GPa}$, which is sufficient to close the cell) compared with data taken outside the diamond anvil cell at ambient pressure conditions.



S3. PRISTINE VS. PRESSURE-CYCLED MATERIALS

Figure S4 summarizes the Raman scattering response of $\text{Cr}_{1/3}\text{TaS}_2$ to pressure cycling. There are small intensity differences due to spot-to-spot variations, but the phonon frequencies are identical. We conclude that there is no hysteresis for the compression pathways that we tested. Although not shown here, $\text{Fe}_{1/3}\text{TaS}_2$ is also reversible upon release of pressure.

Figure S4. Raman scattering response of pristine and pressure-cycled $\text{Cr}_{1/3}\text{TaS}_2$ at room temperature. Data at “pristine” conditions was taken just after the diamond anvil cell was closed. Overall, this system is reversible upon release of the pressure.



S4. TUNABILITY OF THE METAL MONOLAYER EXCITATION

Table S1 summarizes the materials of interest in this work and the characteristics of their metal monolayer excitations. Both in-plane and out-of-plane components are included. The range of linear behavior is also indicated.

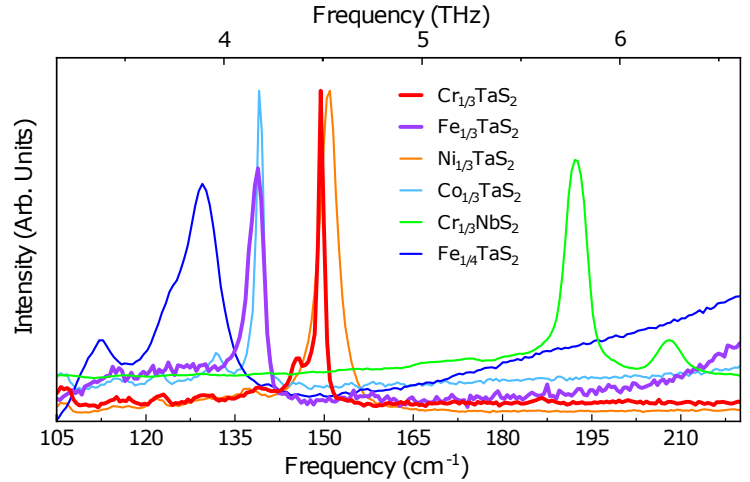
Figure S4 shows a close-up view of the superlattice excitations in this family of materials. A comparison of the metal monolayer excitation in $\text{Cr}_{1/3}\text{TaS}_2$ and $\text{Ni}_{1/3}\text{TaS}_2$ is interesting.

TABLE S1. Summary of the metal monolayer positions and tunability in this class of intercalated chalcogenides. The pressure and strain tunability refer to the in-plane metal monolayer excitations that we study here.

Material	Position of the in- and out-of-plane metal monolayer excitations	Pressure tunability	Strain tunability
$\text{Cr}_{1/3}\text{TaS}_2$	$150 + 172 \text{ cm}^{-1}$ ($4.5 + 5.2 \text{ THz}$)	linear hardening 0 - 17 GPa	linear softening
$\text{Cr}_{1/3}\text{NbS}_2$	$194 + 209 \text{ cm}^{-1}$ ($5.8 + 6.3 \text{ THz}$)	linear hardening 0 - 3 GPa	–
$\text{Fe}_{1/3}\text{TaS}_2$	$139 + 156 \text{ cm}^{-1}$ ($4.2 + 4.7 \text{ THz}$)	linear hardening 0 - 3 GPa	–
$\text{Fe}_{1/4}\text{TaS}_2$	$129 + 112 \text{ cm}^{-1}$ ($3.9 + 3.4 \text{ THz}$)	linear hardening 0 - 2.5 GPa	–

Although they are at similar frequencies, the line widths are very different. We conclude that the Cr analog has a more well-ordered $\sqrt{3} \times \sqrt{3}$ metal monolayer and a longer lifetime.

Figure S5. *Close-up view of the Raman scattering response of the different transition metal intercalated chalcogenides of interest in this work. These spectra were taken at 300 K.*



- [1] Mao, H. K., Bell, P. M., Shaner, J. W., and Steinberg, D. J., Specific volume measurements of Cu, Mo, Pd, and Ag and calibration of the ruby R_1 fluorescence pressure gauge from 0.06 to 1 Mbar, J. Appl. Phys. **49** 3276-3283 (1976).
- [2] Mao, H. K., Xu, J., and Bell, P. M., Calibration of the ruby pressure gauge to 800 kbar under quasi-hydrostatic conditions, J. Geophys. Res. **91**, 4673-4676 (1986).