Supplemental Material
High pressure control of optical nonlinearity in the polar Weyl semimetal TaAs

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I. SHG-RA DATA IN PARALLEL POLARIZATION GEOMETRY

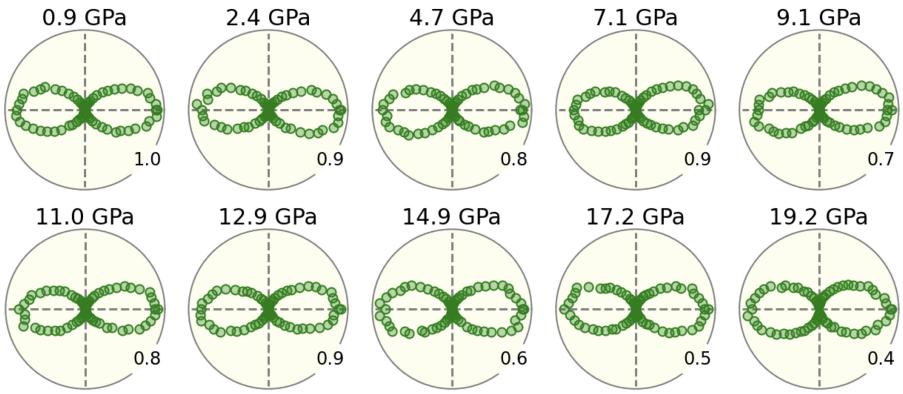

FIG. S1: SHG-RA patterns at select pressure in parallel polarization geometry measured upon compression.

The most general form of the electric-dipole SHG-RA intensity in parallel polarized geometry under normal incidence is given by $I_{\parallel}(\varphi) \propto |B_1 \cos^3(\varphi) + B_2 \cos^2(\varphi)\sin(\varphi) + B_3 \cos(\varphi)\sin^2(\varphi) + B_4 \sin^3(\varphi)|^2$, similar to the case for crossed polarized geometry described in the main text. From the (112) face of TaAs in the tetragonal $4mm$ phase, $B_2 = B_4 = 0$ by symmetry, $B_1 = \gamma(2a^2\chi_{zxx} + 4a^2\chi_{xzx} + c^2\chi_{zzz})$ and $B_3 = \gamma(4a^2 + 2c^2)\chi_{xzx} + (2a^2 + c^2)\chi_{zxx}$, where $\gamma = 1/\left(\frac{2a^3}{c} + ac\sqrt{2 + \frac{c^2}{a^2}}\right)$ and a and c are the TaAs lattice constants.

Figure S1 shows SHG-RA patterns at various pressures measured in parallel polarization geometry. For all pressures, the patterns are dominated by the χ_{zzz} containing B_1 term, which was previously shown to be an order of magnitude larger than both χ_{zxx} and χ_{xzx} [1]. Nonetheless, careful examination of the patterns shows that the nodes at 90° and 270° are lifted at high pressure, evidencing appearance of a B_4 contribution that is only allowed in the hexagonal phase.

II: DEPARTURE FROM $4mm$ POINT GROUP AT AMBIENT PRESSURE

For an ideal tetragonal crystal with point group $4mm$, the A_1 term in $I_{\perp}(\varphi)$ is forbidden by symmetry (see main text) and the pattern should exhibit nodes at 0°, 90°, 180° and 270°. Yet we observe a small but finite A_1 component in our ambient pressure SHG-RA patterns taken outside the DAC , manifested as a lifting of the nodes at 0° and 180°. Our simulation results show that this is not due to misalignment, implying that the crystals may exhibit a slight departure from $4mm$ symmetry.

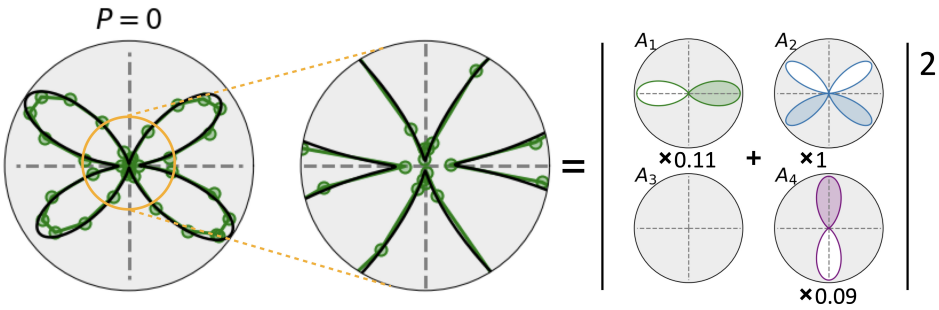

FIG. S2: Typical cross-polarized SHG-RA pattern from TaAs (112) acquired at ambient pressure outside the DAC (left). A zoom-in (middle) shows a lifting of the nodes at 0° and 180°, indicating a finite A_1 term. Fits to the general form for $I_\perp(\varphi)$ are overlaid as black lines. A decomposition of the fits into its $A_1 \rightarrow A_4$ components is shown to the right. Filled versus empty lobes represent opposite signs of the associated trigonometric function. The fitted amplitude of the various terms is shown below, which are normalized to A_2 for $P < P_c$.

III: PRESSURE DEPENDENCE OF A_1 AND A_4

The SHG-RA patterns measured in crossed polarized geometry were fit to $I_\perp(\varphi) \propto |A_1 \cos^3(\varphi) + A_2 \cos^2(\varphi)\sin(\varphi) + A_3 \cos(\varphi)\sin^2(\varphi) + A_4 \sin^3(\varphi)|^2$. The fitted amplitudes of A_2 and A_3 are shown in the main text. Figure S3 shows the pressure dependence of the fitted amplitudes of A_1 and A_4. Subtle kinks appear in $|A_1|$ and $|A_4|$ around the critical pressure P_c. However, as expected, $|A_4|$ does not change as drastically as $|A_2|$ because A_4 does not contain χ_{zzz}. Also, as expected, $|A_1|$ does not exhibit a clear a kink at P_c as $|A_3|$ because A_1 is present in both the low and high pressure phases.

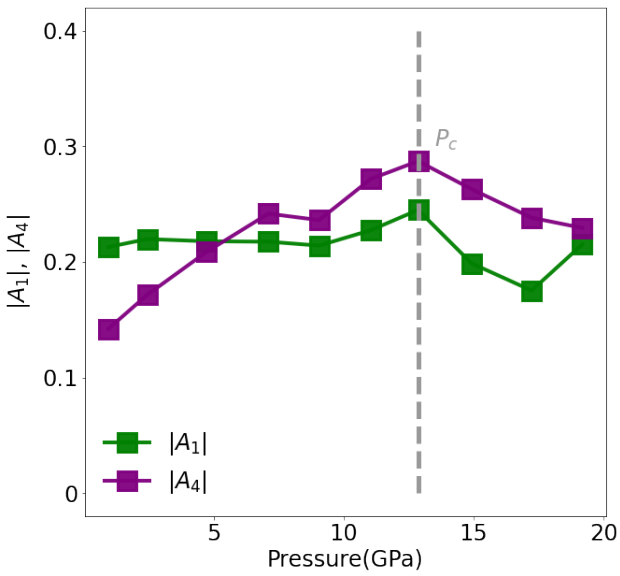

FIG. S3: Pressure dependence of the fitted amplitudes of A_1 (green) and A_4 (purple) normalized to the maximum value of A_2 for $P < P_c$

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[1] Wu, L. *et al.* Giant anisotropic nonlinear optical response in transition metal monopnictide Weyl semimetals. *Nature Physics* **13**, 350–355 (2017). URL https://www.nature.com/articles/nphys3969. Number: 4 Publisher: Nature Publishing Group.