Optical second harmonic susceptibility in the Weyl semimetal WTe_2

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Overview

- Studies on TaAs, a Weyl semimetal showed unusually large second-order nonlinear optical susceptibility.[1]
- Subsequently, models that connects the band-structure geometry and the nonlinear susceptibility were established to explain this anomaly.
- Second harmonic generation in the Weyl semimetal, tungsten telluride (WTe_2).

[1] Wu, L. et al., Giant anisotropic nonlinear optical response in transition metal monopnictide Weyl semimetals, *Nat. Phys.* **13**, 350-355 (2017)

Polarization response in nonlinear media

- The nonlinear optical **polarization response**, P(t) of a medium is given by,
- $P_i = \epsilon_0 \chi^{(1)}_{ij} E_j + \epsilon_0 \chi^{(2)}_{ijk} E_j E_k + \epsilon_0 \chi^{(3)}_{ijkl} E_j E_k E_l + \dots$
- $\chi^{(2)}_{ijk}$ transforms as a third-rank tensor

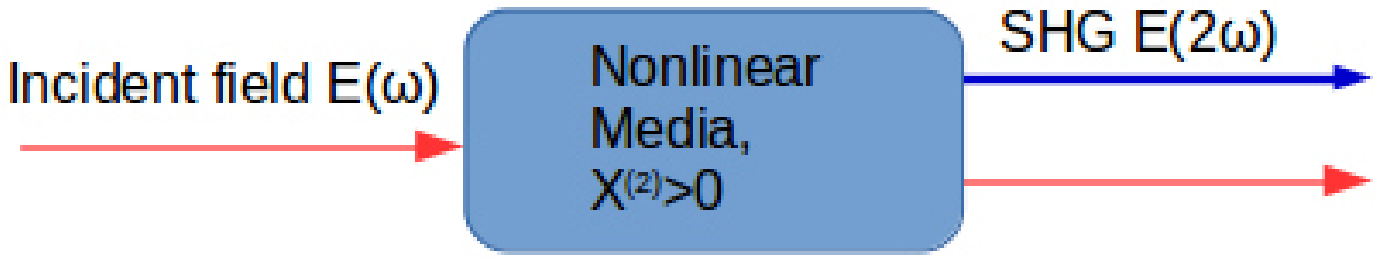
- For an electric field, $E(t) = (Ee^{-i\omega t} + c.c)$ and real $\chi^{(2)}$,

Second Harmonic Generation

$$\mathrm{P}^{(2)}(t) = (\epsilon_0 \chi^{(2)} E^2 e^{-i\,2\omega t} + c.c) + 2\epsilon_0 \chi^{(2)} E E^*$$

Influence of Inversion Symmetry on the Second-Order Nonlinear Response

- Under inversion,
  $\mathscr{P}$E = - E,
  $\mathscr{P}$P = - P,
  Therefore,
  $P_i^{(2)} = \epsilon_0 \chi_{ijk}^{(2)} E_j E_k \neq 0$ iff
  $\mathscr{P}\chi_{ijk}^{(2)} \neq \chi_{ijk}^{(2)}$
  $P^{(2)} \neq 0 \implies$ broken inversion



Morphology of WTe_2 xtal

- Orthorhombic : C_{2v}(mm2).
- Symmetry elements : C_{2z}, $\sigma_v(xz)$, $\sigma_v(yz)$.
- Hence by Neumann's principle,
  the nonzero dipole-$\chi^{(2)}_{ijk}$: xxz, xzx, yyz, yzy, zyy, zzz

Second Harmonic Generation and crystal orientation

- The components of the susceptibility tensor is defined as,

$$P_i^{(2)}(2\omega) = \epsilon_0 \sum_{jk} \chi_{ijk}^{(2)} E_j(\omega) E_k(\omega)$$

$$\begin{bmatrix} E_x^{2\omega} \\ E_y^{2\omega} \\ E_z^{2\omega} \end{bmatrix} \propto \begin{bmatrix} P_x^{(2)} \\ P_y^{(2)} \\ P_z^{(2)} \end{bmatrix} = \epsilon_0 \begin{bmatrix} (\chi_{xxz} + \chi_{xzx}) \mathsf{E}_x \mathsf{E}_z \\ (\chi_{yyz} + \chi_{yzy}) \mathsf{E}_y \mathsf{E}_z \\ \chi_{zxx} \mathsf{E}_x^2 + \chi_{zyy} \mathsf{E}_y^2 + \chi_{zzz} \mathsf{E}_z^2 \end{bmatrix}$$

- Applied field along z-axis, NO SHG.
- Applied field along x-axis or y-axis, SHG exist.

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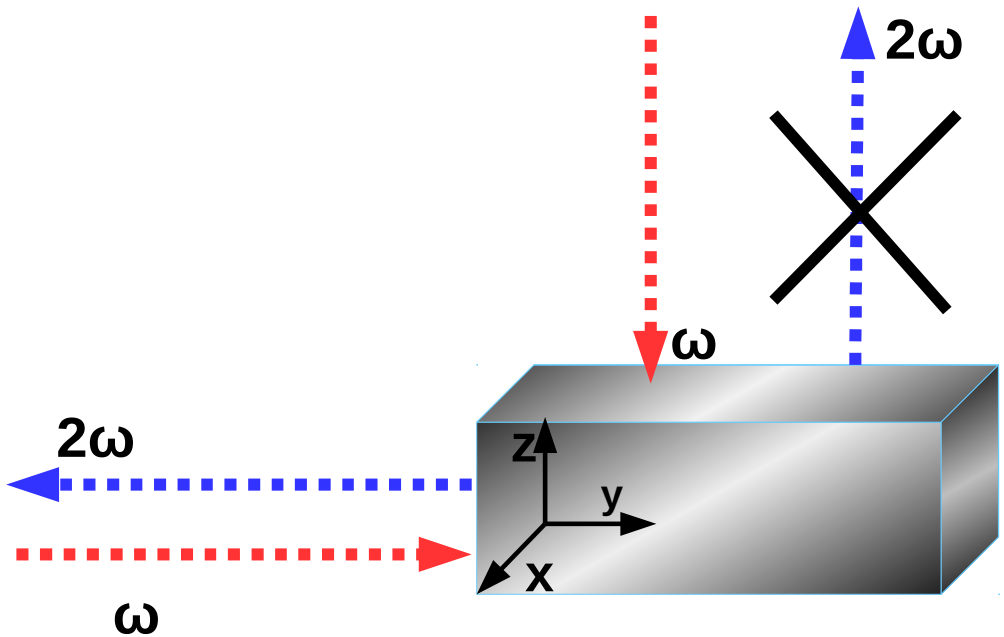
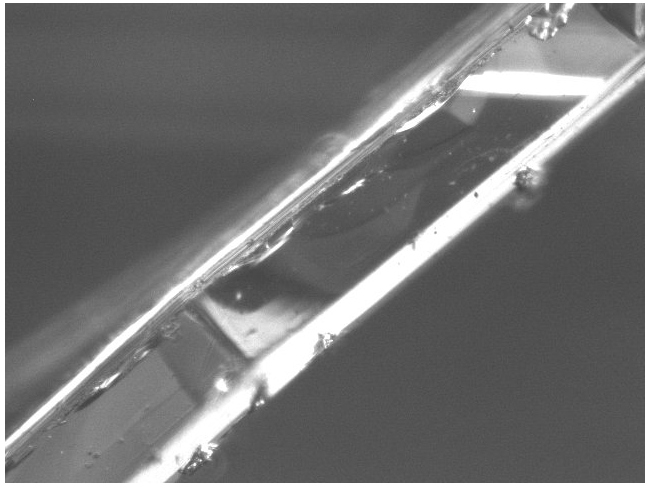
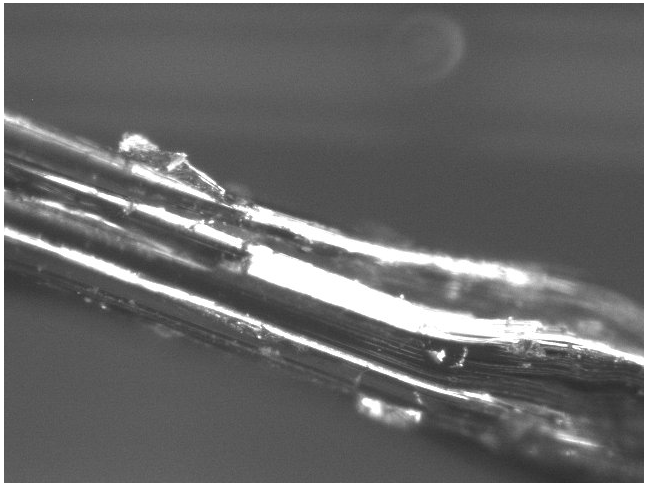
Second Harmonic Generation in WTe_2 xtal



Second Harmonic Generation and crystal orientation

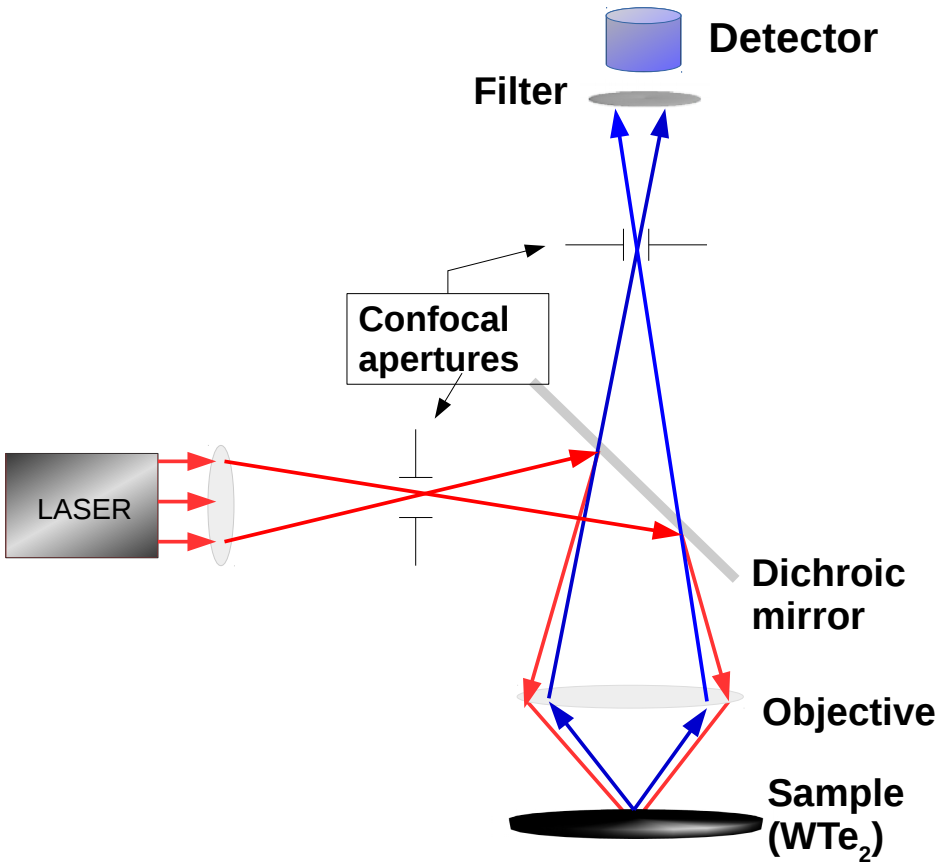


ab face
(perpendicular to z-axis)

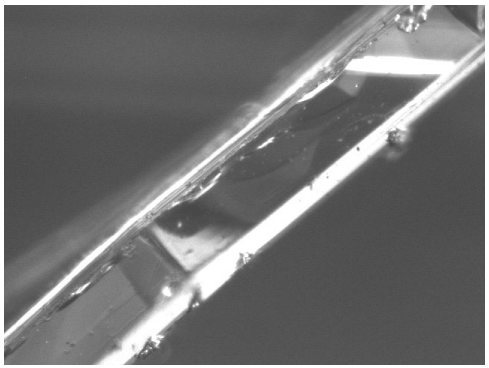


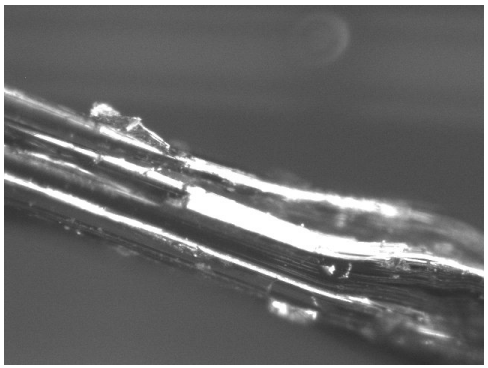
ac face
(perpendicular to x-axis)

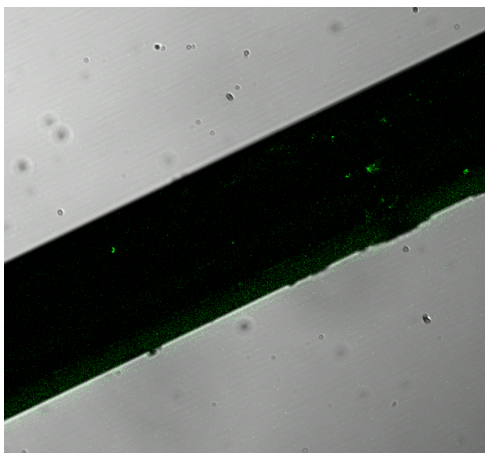
Microscope SHG measurement setup

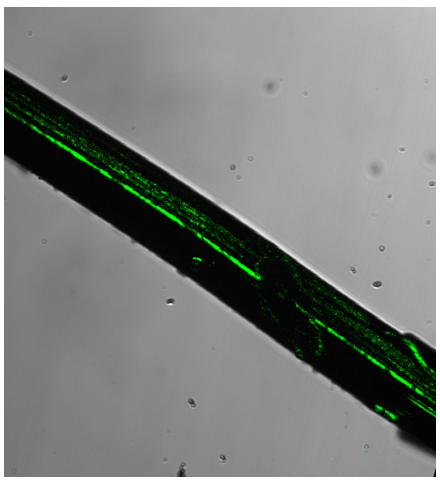


SHG from edge and face





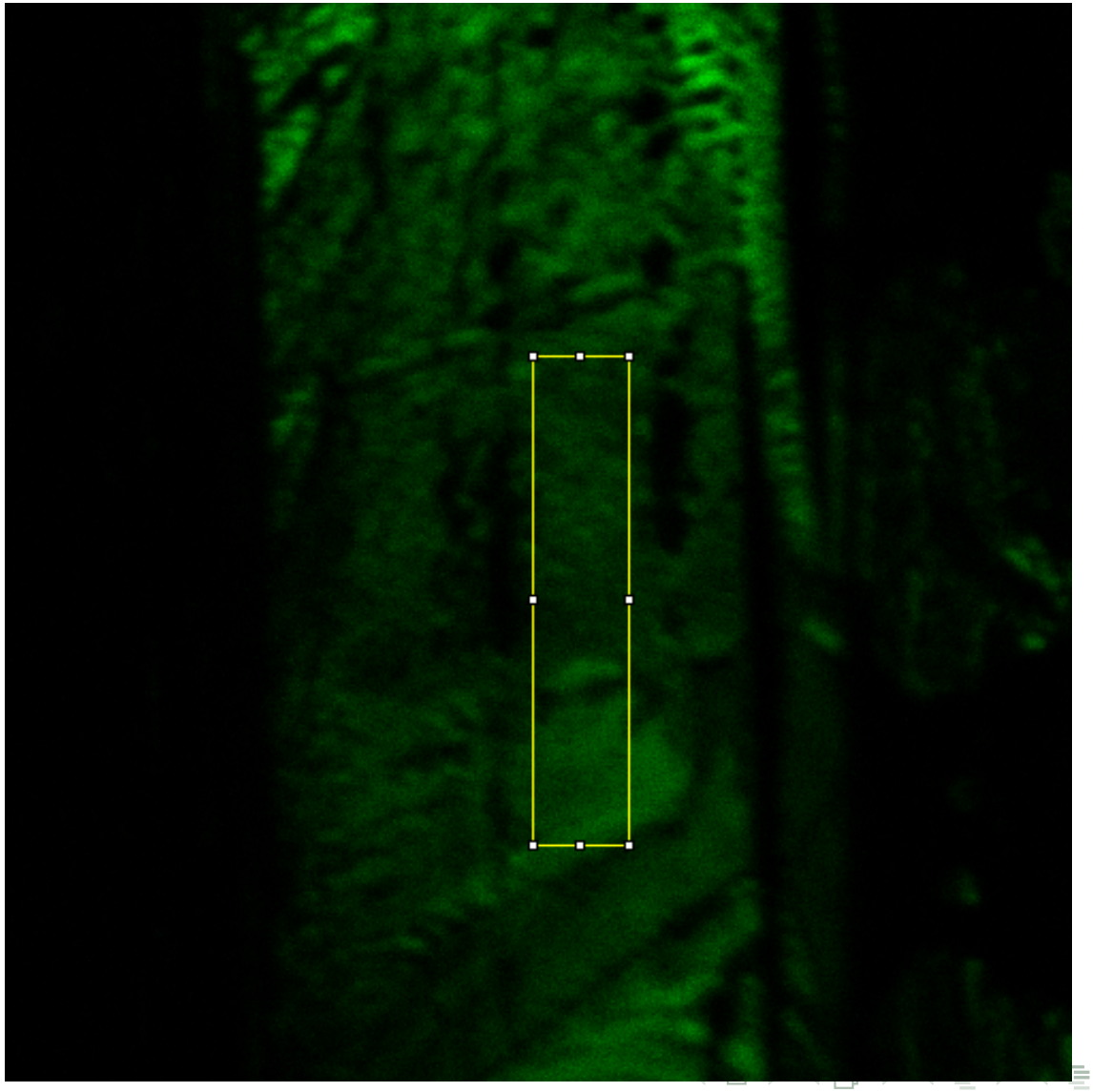




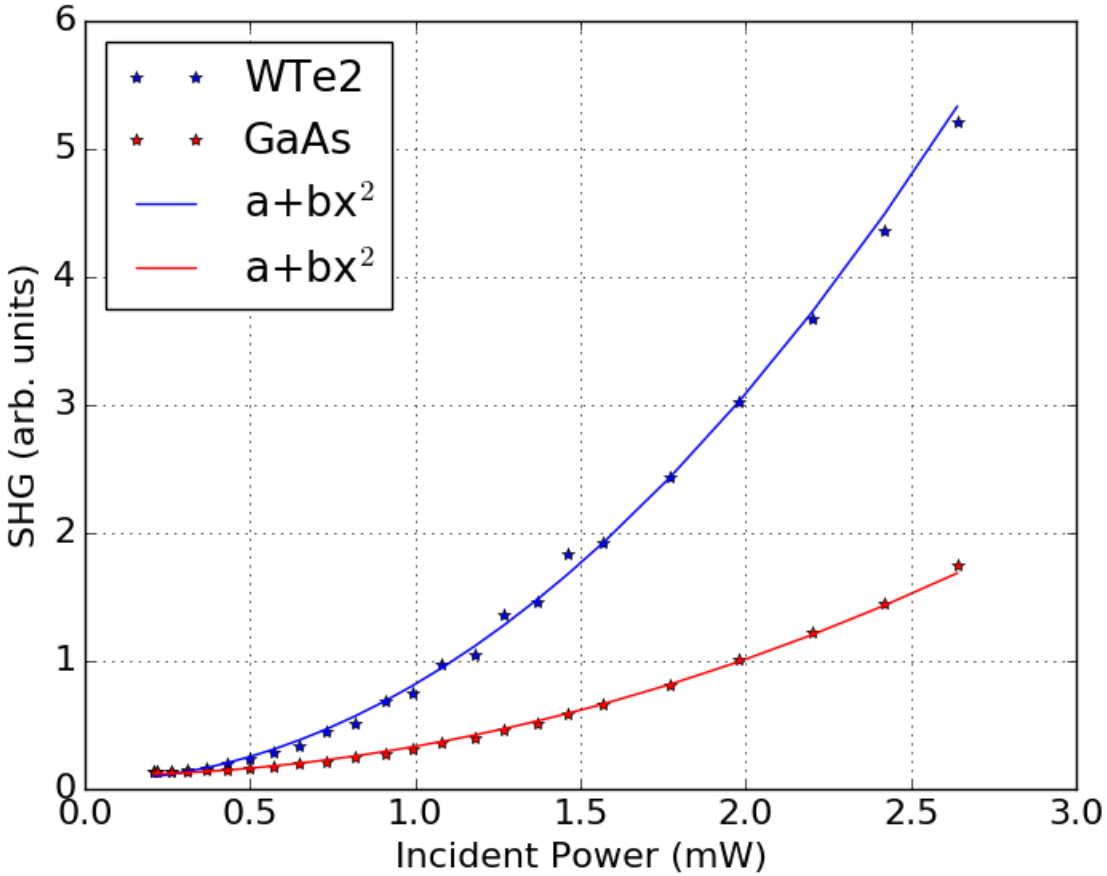
ab face - No SHG

ac face - SHG

SHG from the acquired images

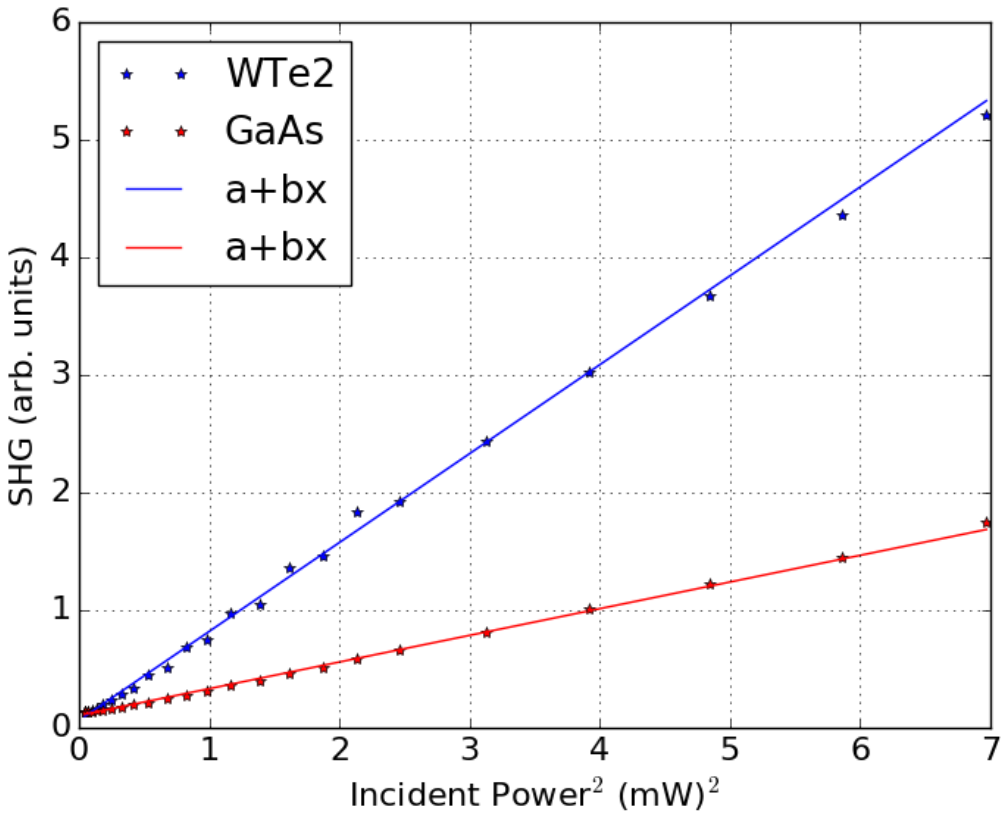


Quadratic power-law behaviour in WTe_2 and GaAs



Quadratic power-law behaviour in WTe_2 and GaAs

$$P_{2\omega} = \frac{8\omega^2}{\epsilon_o c^3 n_\omega^3} \frac{L^2}{A} \chi_{eff}^{(2)2} P_\omega^2$$



$$\frac{\chi_{WTe_2}^{(2)}}{\chi_{GaAs}^{(2)}} \approx 2$$

Summary

- Second harmonic generation in the Weyl semimetal WTe_2.
- SHG is observable only for light reflected from the edges of the crystal, not from the natural planar surface of WTe_2.
- Hence the measurement has to be done in a microscope.
- The $\chi^{(2)}$ of WTe_2 is greater than that of GaAs.
- We are in the process of doing a polarization study to determine the individual $\chi^{(2)}_{ijk}$