Nonlinear optical and terahertz spectroscopy of topological semimetals

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Part 1 :band geometry

- **Introduction to nonlinear optics and a new perspective on band geometry**

- **Discovery of the largest second harmonic generation (SHG) in polar Weyl semimetals TaAs**

Acknowledgements

Joe Orenstein (UCB)

Shreyas Patankar (UCB)

Darius Torchinsky (Temple)

Joel Moore (UCB)

Takahiro Morimoto (UCB)
Adolfo Grushin (UCB)
Dan Parker (UCB)
James Analytis (UCB)
Nityan Nair (UCB)

Motivation

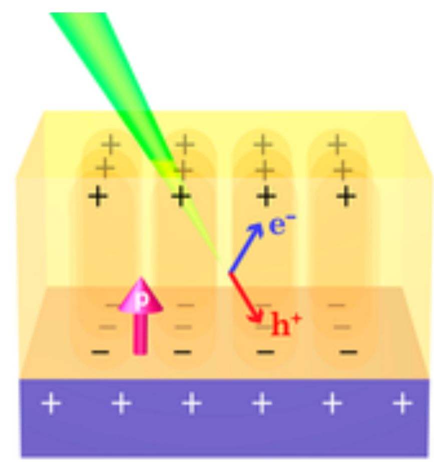
Topology ⟺ Nonlinear optical effects

- Quantum Hall effect
- Topological insulators
- Weyl semimetal

- Photovoltaics
- Second harmonic generation

Quantized Hall conductance
(von Klitzing, *PRL*, 80; Chang, Xue. *Science*, 2013)
Quantized Terahertz Faraday & Kerr rotation
(Wu, Armitage. ***Science*** 2016)

$J_i(0) \propto \sigma^{(2)} I(\omega) \rightarrow$ “bulk photovoltaic effect”



Topological nonlinear optics?

Guidance to find better photovoltaics based on topological materials ?

Part 1

- **Introduction to nonlinear optics and a new perspective on band geometry**

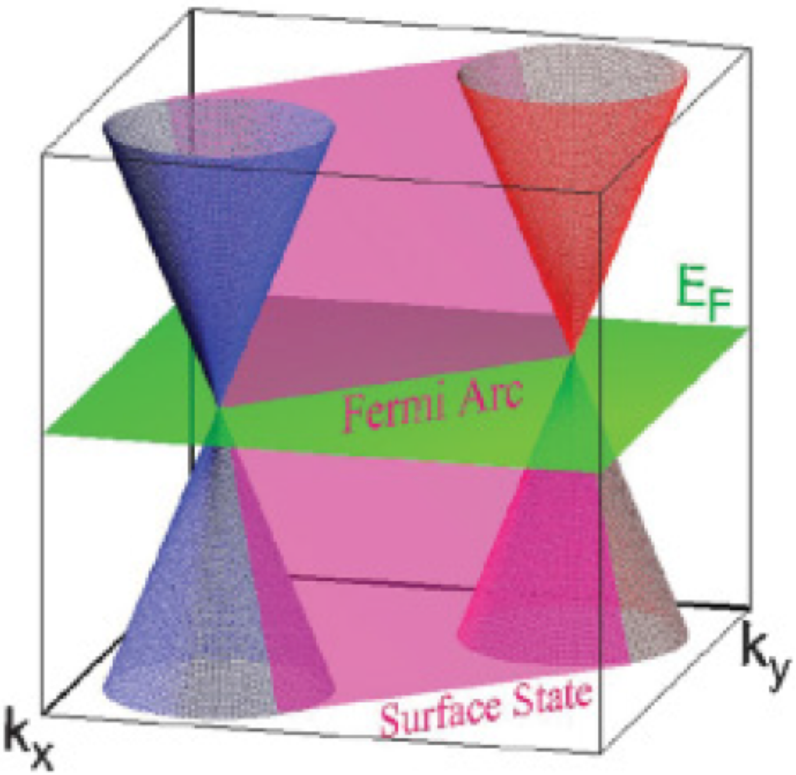
- **Discovery of the largest second harmonic generation (SHG) in polar Weyl semimetals TaAs**

Weyl semimetals (WSMs)

PHYSICAL REVIEW B **83**, 205101 (2011)

Topological semimetal and Fermi-arc surface states in the electronic structure of pyrochlore iridates

Xiangang Wan,[1] Ari M. Turner,[2] Ashvin Vishwanath,[2,3] and Sergey Y. Savrasov[1,4]



$$H(\mathbf{k}) = \varepsilon_0\sigma_0 \pm \hbar v_F(\mathbf{k} - \mathbf{k}_0)\cdot\boldsymbol{\sigma}$$

(H. Weyl 1929)

"3D graphene"

Broken Inversion/Time Reversal Symmetry

Murakami. ***New. J. Phys.*** (2007)
Wan, *et al*. ***PRB.*** (2011)

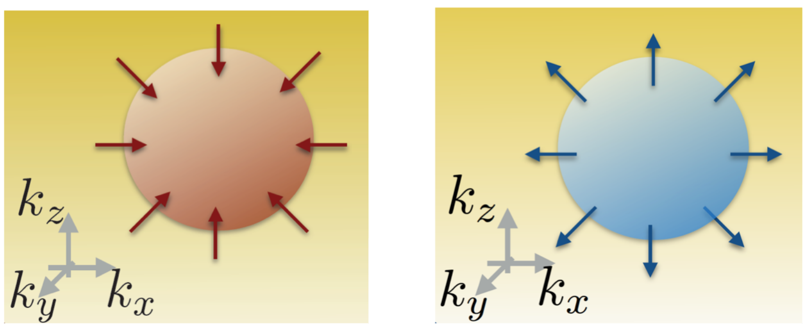
Band topology in WSMs

Berry curvature

$$\boldsymbol{A} = -i\langle u_k | \nabla_k u_k \rangle$$

$$\boldsymbol{\Omega}(\mathbf{k}) = \nabla_k \times \boldsymbol{A}(\mathbf{k})$$

Think of like magnetic field living in k-space (Monopoles)

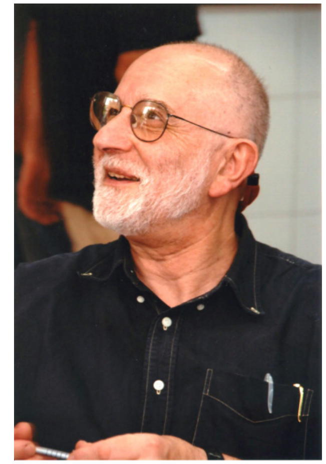


$$\frac{1}{2\pi}\oint_{FS} \Omega(\mathbf{k})d\mathbf{k} = C_i$$

Weyl points are stable unless they meet in momentum space and annihilate each other.

Berry (geometrical) phase

Sir Michael Berry (1984)



Nonzero Berry curvature requires breaking of either Inversion or Time-reversal symmetry !

Inversion symmetry

$$\Omega(\mathbf{k}) = \Omega(-\mathbf{k})$$

Time-reversal symmetry

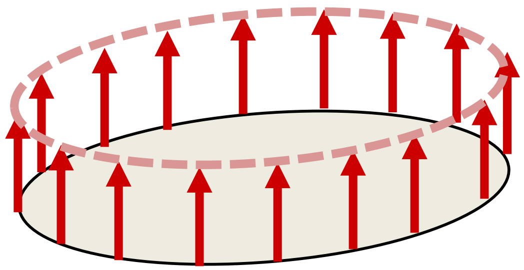
$$\Omega(\mathbf{k}) = -\Omega(-\mathbf{k})$$

With Both symmetries

$$\Omega = 0$$

T and I breaking allow new optical phenomena.

Broken T symmetry



$$\langle \mathbf{\Omega}(\mathbf{k}) \rangle \neq 0$$

$$\Omega_n(-k) = \Omega_n(k)$$

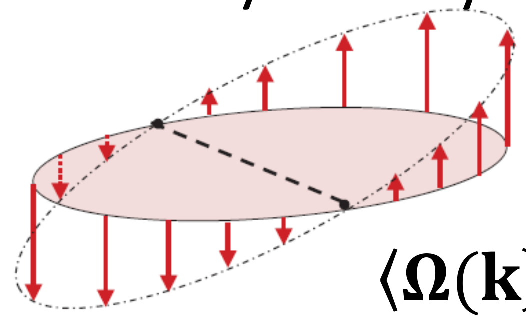
$$j_x = \sigma_{xy} E_y$$

Intrinsic Anomalous Hall effect (DC transport)

Faraday and Kerr rotation without applied B

$$\sigma_{xy} = \frac{e^2}{\hbar} \int_{\mathrm{BZ}} \frac{d^2k}{(2\pi)^2} \Omega_{k_x k_y}$$

Broken I symmetry



$$\langle \mathbf{\Omega}(\mathbf{k}) \rangle = 0$$

$$\Omega_n(-k) = -\Omega_n(k)$$

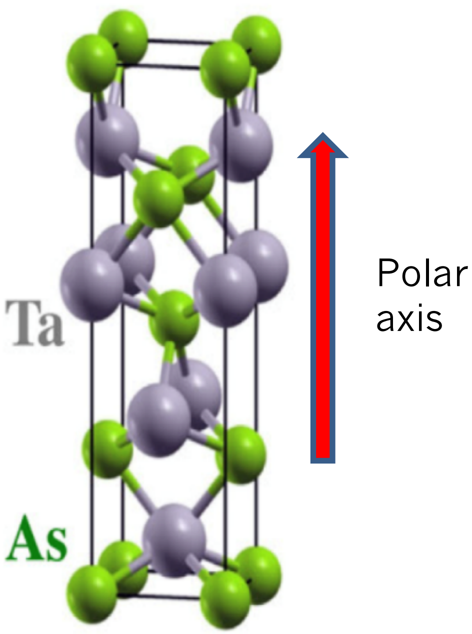
$$J_i = \sigma_{ijk} E_j E_k$$

Second order nonlinearity

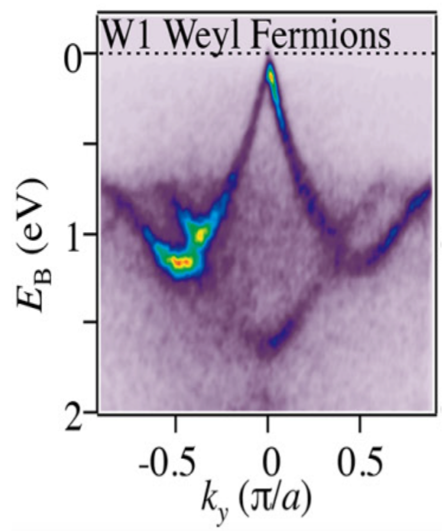
Zoo of acronymic effects!
SHG, DFG, **CPGE**, LPGE, etc.

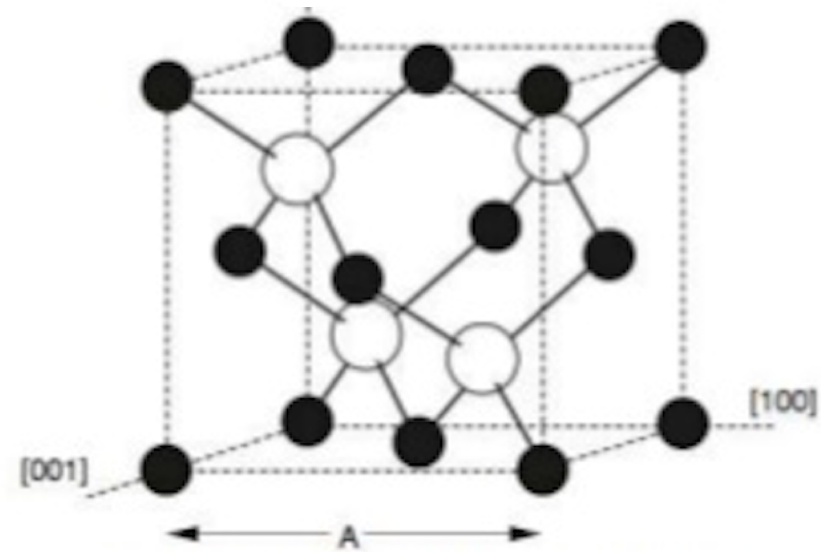
Material realization---Transition metal monopnictides

WSMs TaAs, TaP, NbAs, NbP break inversion and are polar metals (or ferroelectric metals) !



$4mm$ point group





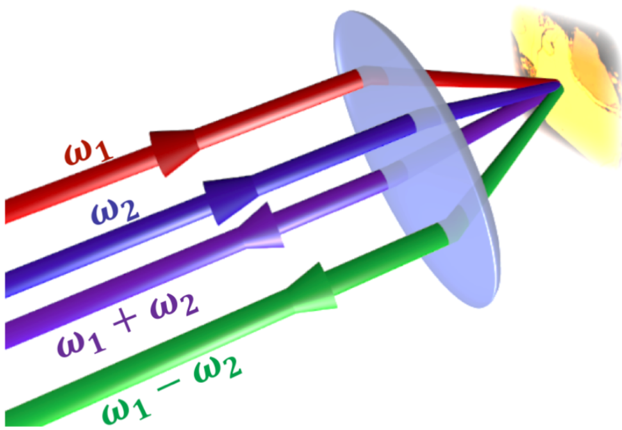
Weng *et al.*, ***PRX*** (2015)
Huang, *et al.*, ***Nat. Comm.*** (2015)
Xu, *et al*, ***Science*** (2015)
Lv, *et al*, ***PRX*** (2015)

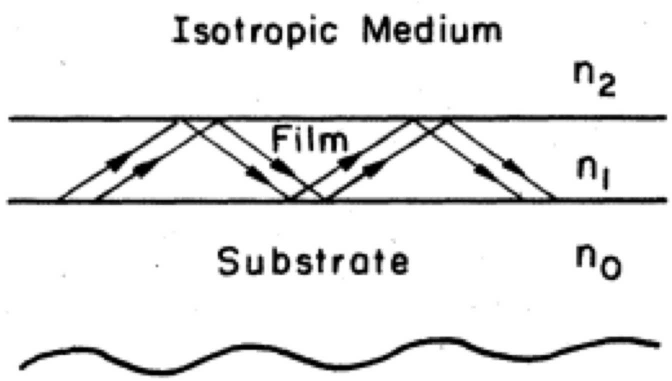
GaAs breaks inversion, but is not polar.

Are there new/enhanced transport and optics effects in inversion-breaking WSMs associated Berry monopoles?

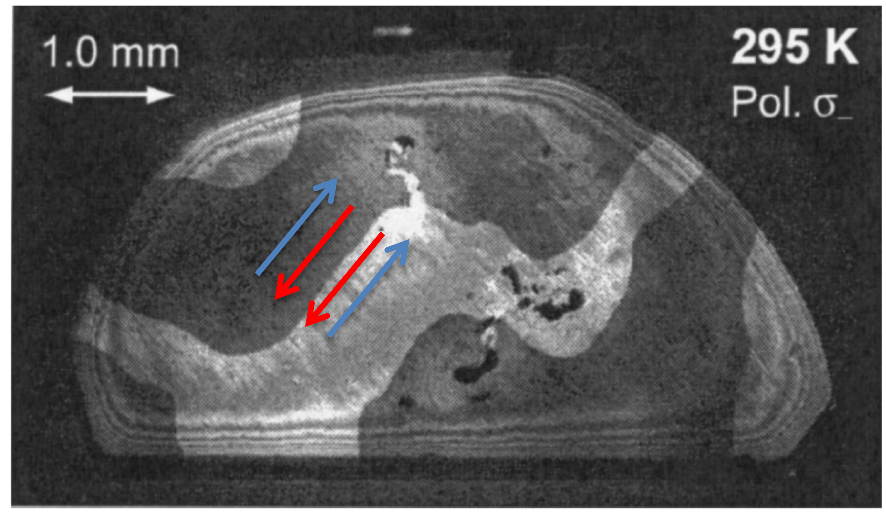
Past nonlinear optics

Focused on probing light conversion and symmetry breaking.





Y. R. Shen, *RMP* (1976)



Cr_2O_3

Fiebig, et al. *JOSA* (2005)

Why nonlinear optics experiments on WSMs?

How textbooks calculate nonlinear optical susceptibility....

$$\chi^{(2)}_{\mu\alpha\beta}(-2\omega,\omega,\omega)$$

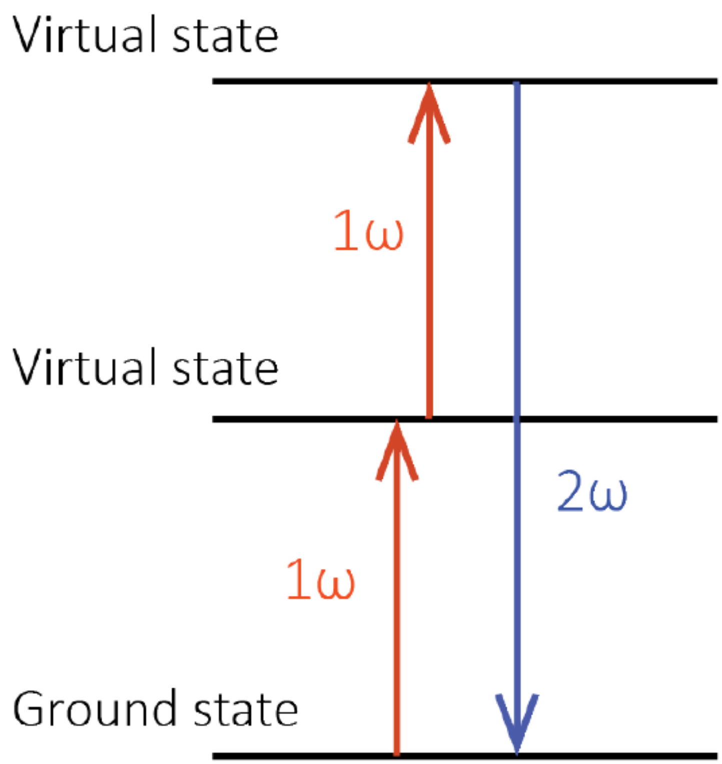
$$= -i\frac{1}{32\varepsilon_0}\left(\frac{e}{m_0\pi\omega}\right)^3 \sum_{n,n',n''}\int_{\mathrm{BZ}} d^3\mathbf{k}$$

$$\times f_{n\mathbf{k}}\left\{\frac{p^{\mu}_{nn'}p^{\alpha}_{n'n''}p^{\beta}_{n''n}+p^{\mu}_{nn'}p^{\beta}_{n'n''}p^{\alpha}_{n''n}}{[E_{n'n}(\mathbf{k})-2\hbar\omega][E_{n''n}(\mathbf{k})-\hbar\omega]}\right.$$

$$+\frac{p^{\alpha}_{nn'}p^{\mu}_{n'n''}p^{\beta}_{n''n}+p^{\beta}_{nn'}p^{\mu}_{n'n''}p^{\alpha}_{n''n}}{[E_{n'n}(\mathbf{k})+\hbar\omega][E_{n''n}(\mathbf{k})-\hbar\omega]}$$

$$\left.+\frac{p^{\beta}_{nn'}p^{\alpha}_{n'n''}p^{\mu}_{n''n}+p^{\alpha}_{nn'}p^{\beta}_{n'n''}p^{\mu}_{n''n}}{[E_{n'n}(\mathbf{k})+\hbar\omega][E_{n''n}(\mathbf{k})+2\hbar\omega]}\right\},$$

$$\mu,\alpha,\beta\in\{x,y,z\}.$$



Why nonlinear optics experiments on WSMs?

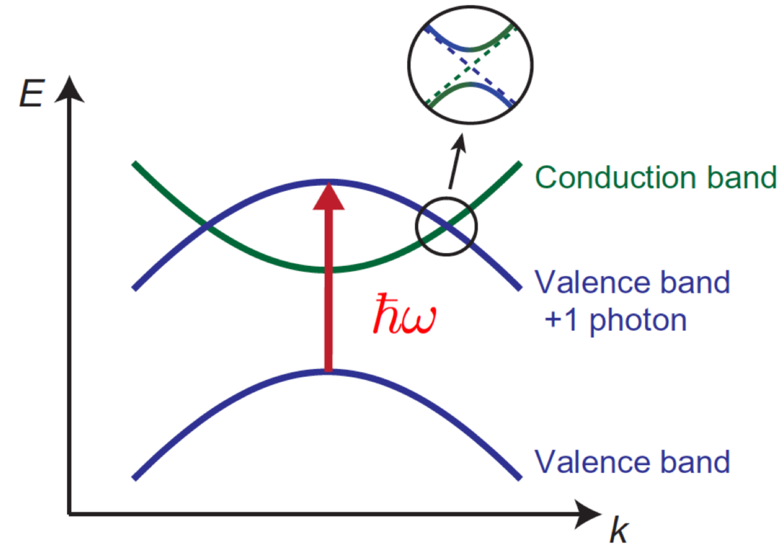
--- Probing Berry Connection & band topology

$$\boldsymbol{a}_n(\boldsymbol{k}) = -i\langle u_{n\boldsymbol{k}}|\nabla_{\boldsymbol{k}} u_{n\boldsymbol{k}}\rangle$$

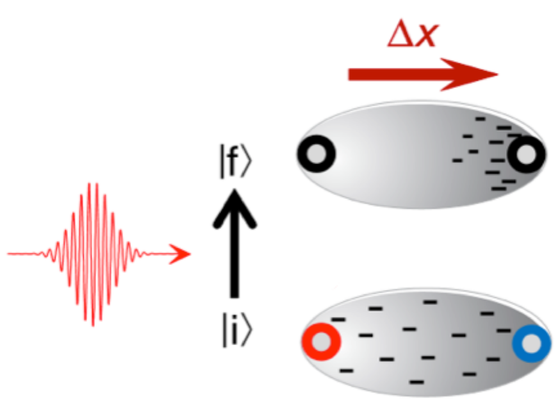
$$\varphi_{12} = \mathrm{Im}(\log v^0_{12})$$

$$R_k = \left[\frac{\partial\varphi_{12}}{\partial k} + a_1 - a_2\right]$$

"Shift vector" measures the change of **intracell coordinates in the transition between the initial and final states.**



Von Baltz ***PRB*** (1979) (1981) ; Sturman & Fridkin & Belinicher ***SPU*** (1980) (1992);
Sipe & Shkrebtii ***PRB.*** (2000); Young & Rappe ***PRL*** 2012;Morimoto & Nagaosa ***Sci. Adv.*** (2016)



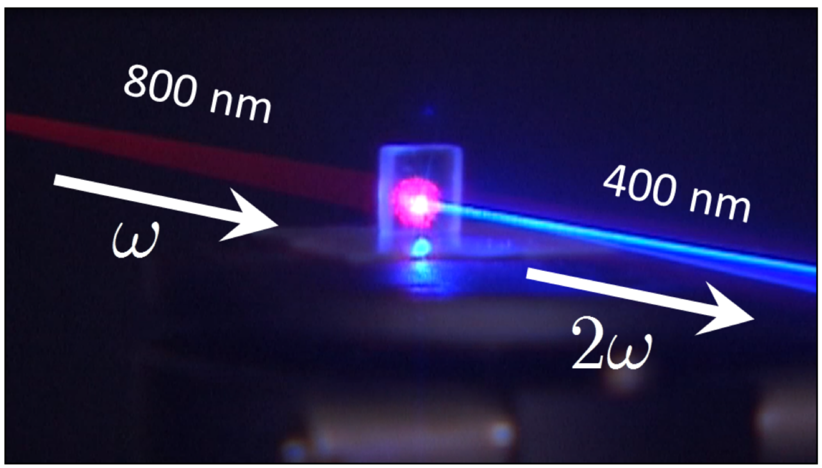
$$\mathrm{Re}\{\sigma^{(2)}_{zzz}(\omega, 0)\} \cong \frac{\pi e^3}{2\hbar\omega^2}\int \frac{d^3\boldsymbol{k}}{(2\pi)^3} |v_{z,12}|^2 R_{zz}(\boldsymbol{k})$$

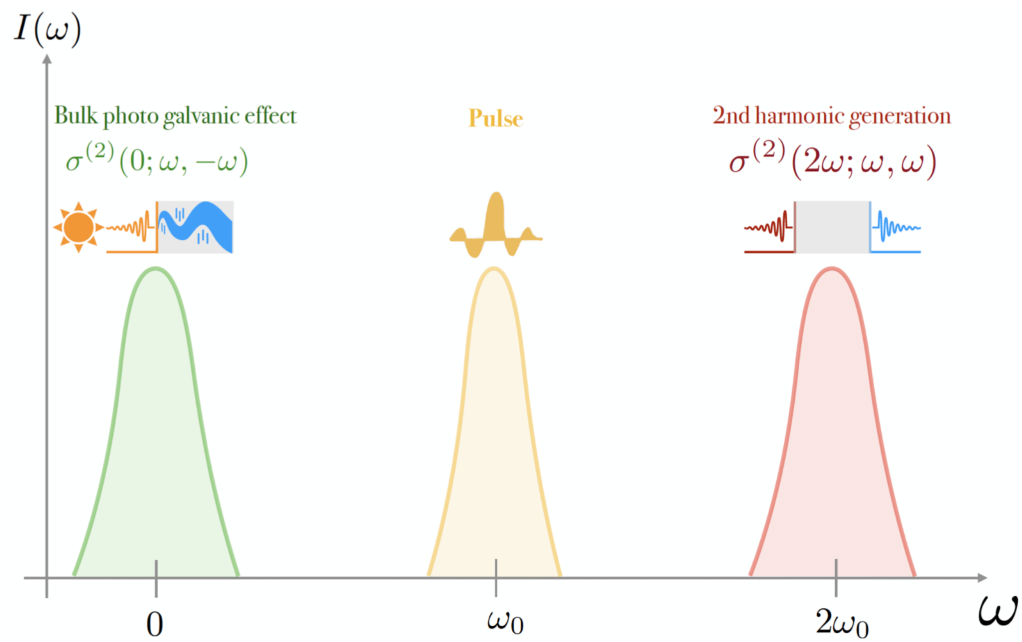
$$\times\left[-\delta(\epsilon_{21} - \hbar\omega) + \quad\right]$$

2^{nd} order nonlinear optical effect in general

$$J_i(\omega_1 \pm \omega_2) = \sigma_{ijk}(\omega_1 \pm \omega_2)E_j(\omega_1)E_k(\omega_2)$$

sum and **difference** frequency generation





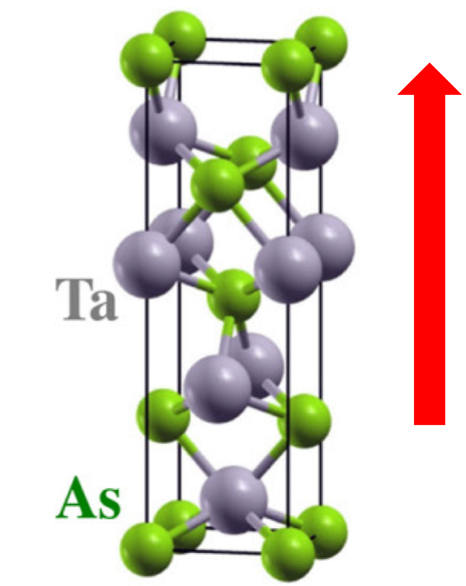
Second harmonic generation (SHG)

In materials without inversion symmetry,

$$\mathbf{P} = \mathbf{P}_0 + \epsilon_0 \chi_e \mathbf{E} + \epsilon_0 \chi^{(2)} \mathbf{E}^2 + \cdots$$

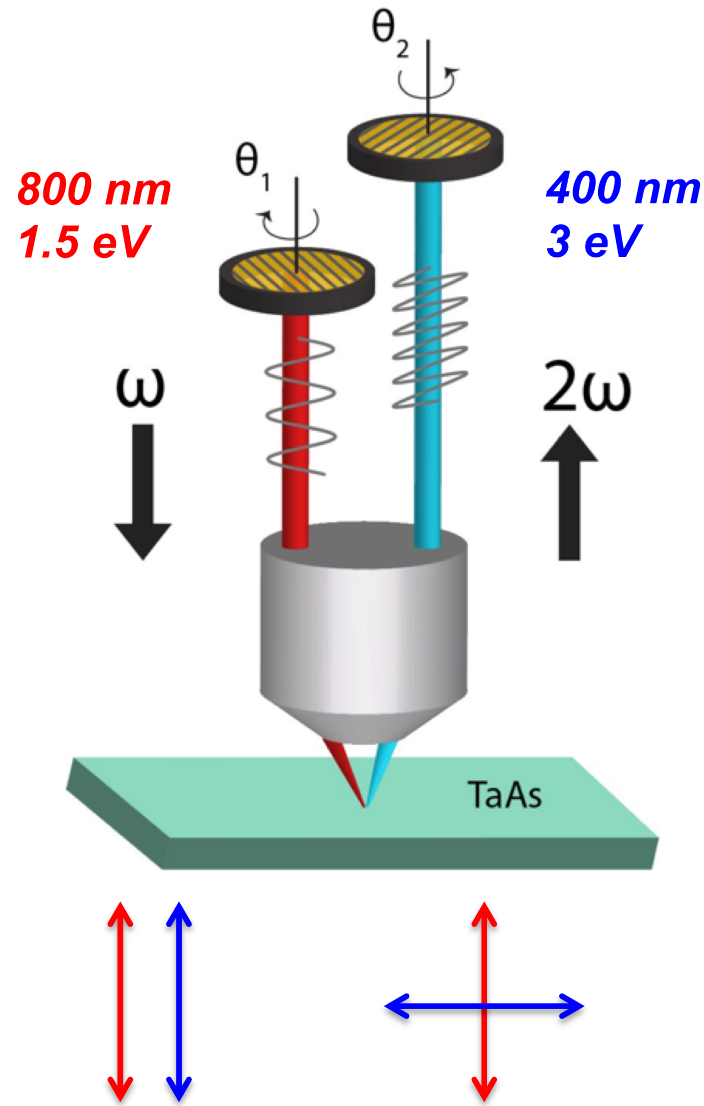
$$P_i(2\omega) = \epsilon_0 \chi_{ijk}(2\omega) E_j(\omega) E_k(\omega)$$

Bloembergen & Pershan. ***Phys. Rev*** (1962)



Polar Axis [001]

$$\chi_{zzz}$$



Parallel **Perpendicular**

Mirror plane *xz* & *yz*, but not *xy*

$4mm$ point group determines **three** non-zero χ_{ijk}.

Part 1

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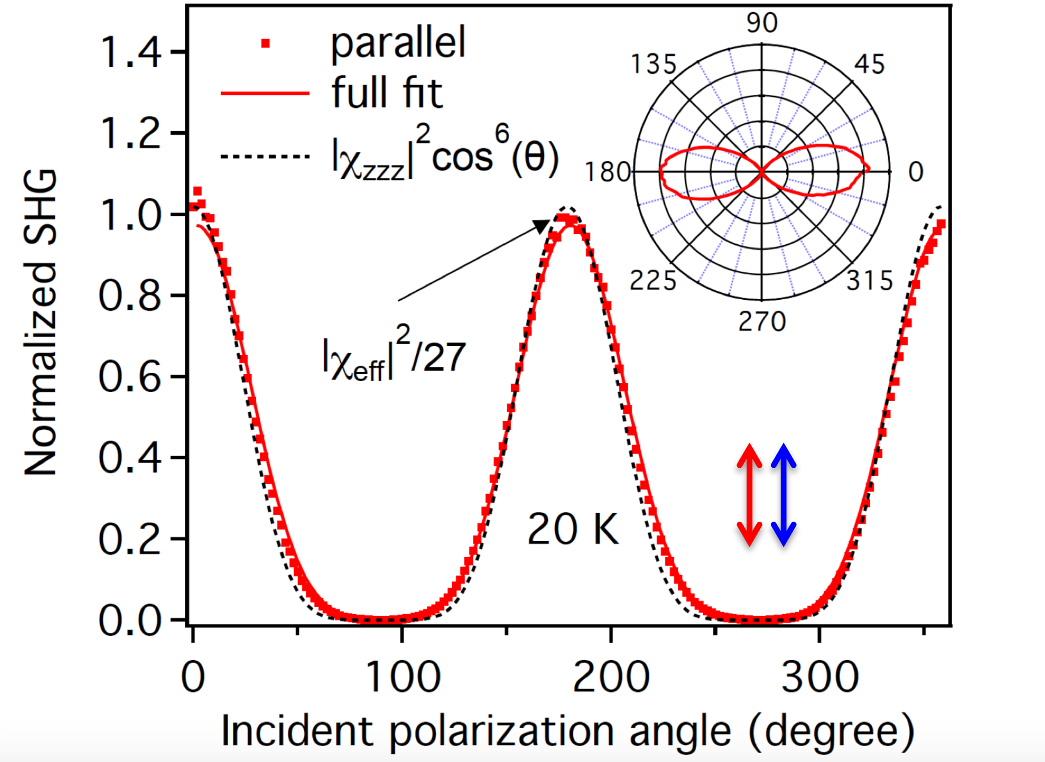
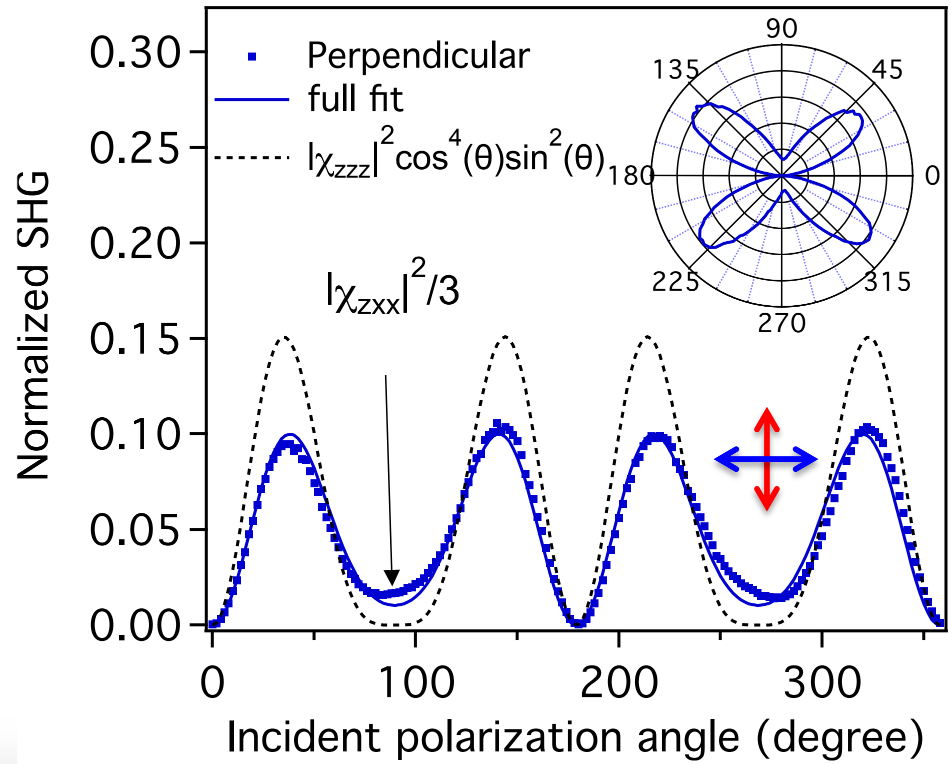
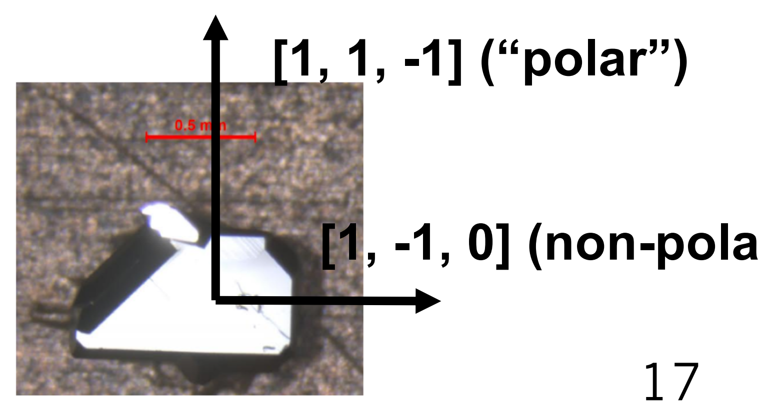
SHG on TaAs (112) surface





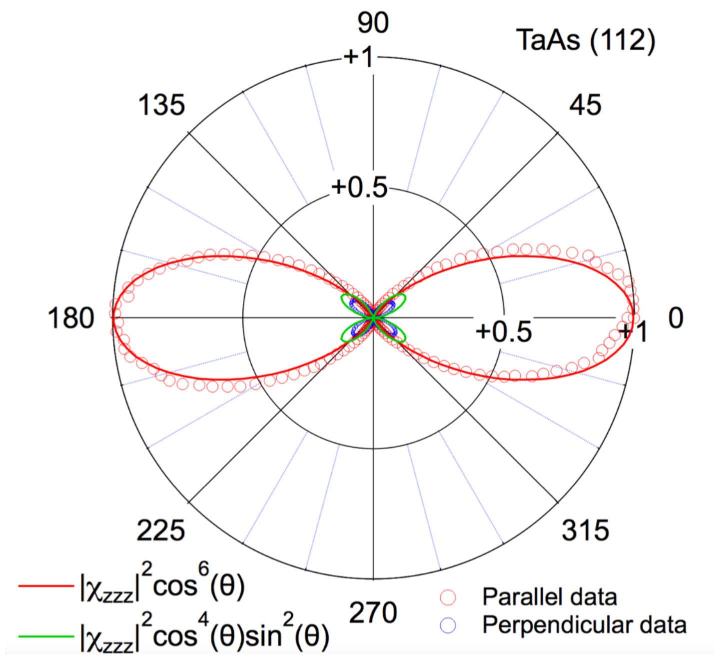
Dominating χ_{zzz} . (effectively 1D-like)

Anisotropy $\chi_{zzz}/\chi_{zxx}, \chi_{zzz}/\chi_{xzx}$ 30-100 in TaAs !
Materials with same χ_{ijk} tensor have anisotropy factor 1-2. e.g. $LiNbO_3$, $BaTiO_3$

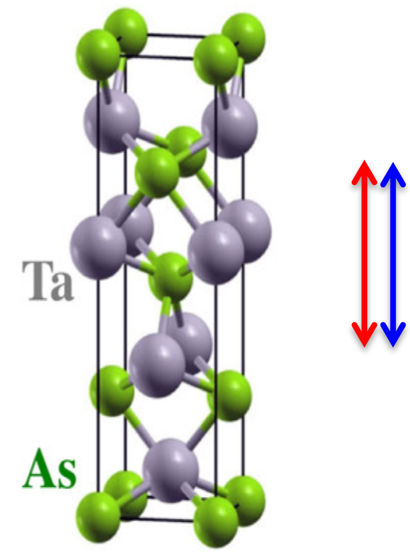


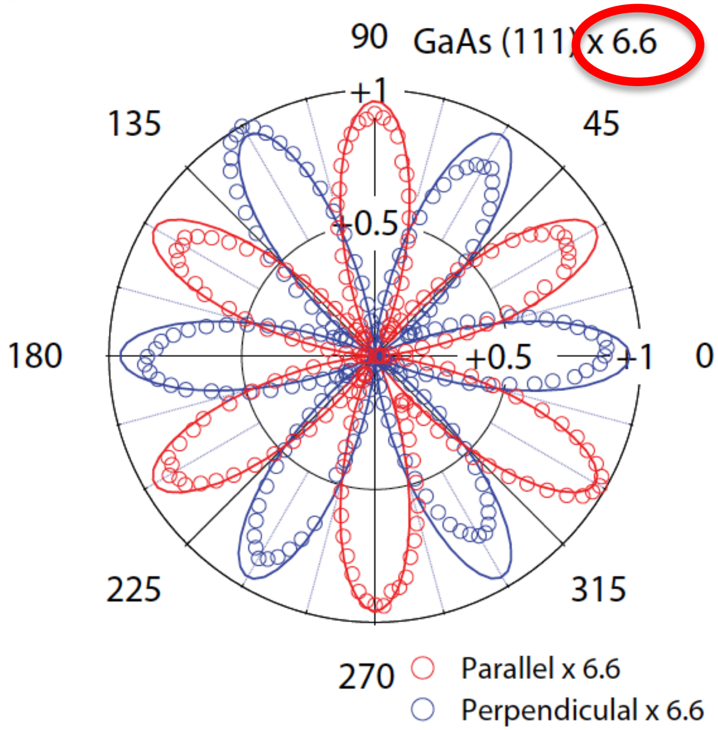
Wu, *et al*. ***arXiv:1609.04894***
Nat. Phys. (2016)

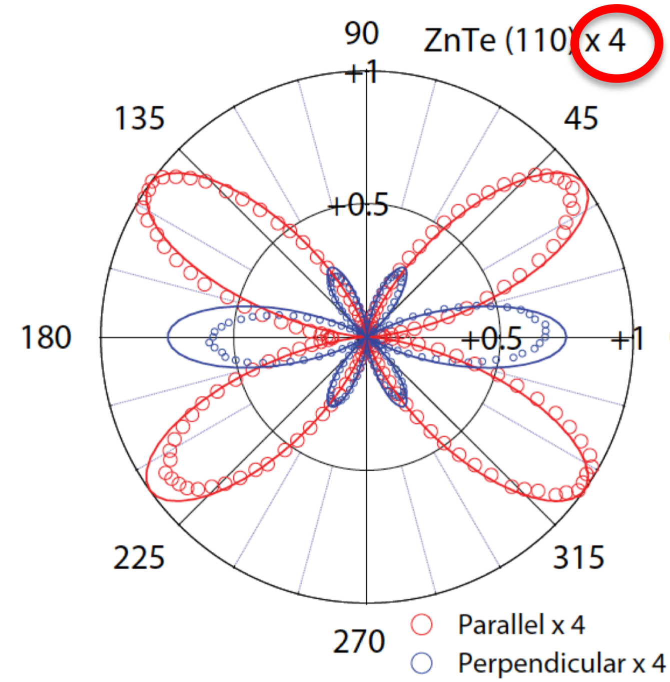
Largest SHG in existing materials



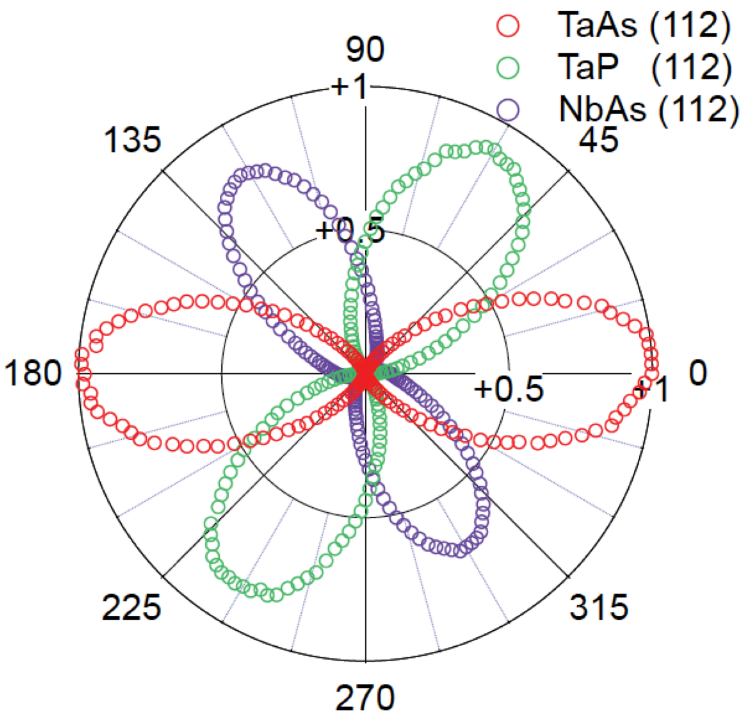
TaAs (100) will have SHG intensity >100 times bigger than GaAs (111)!







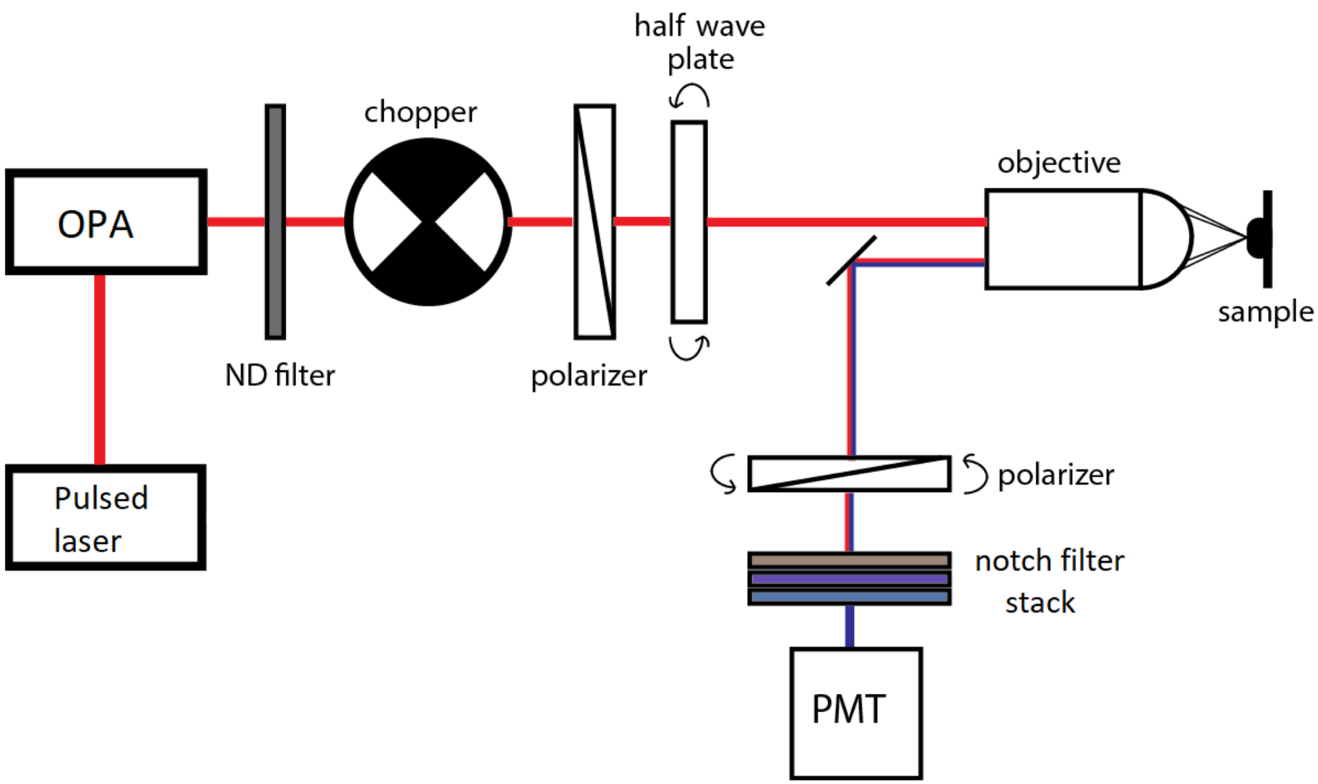
Other WSMs TaP & NbAs



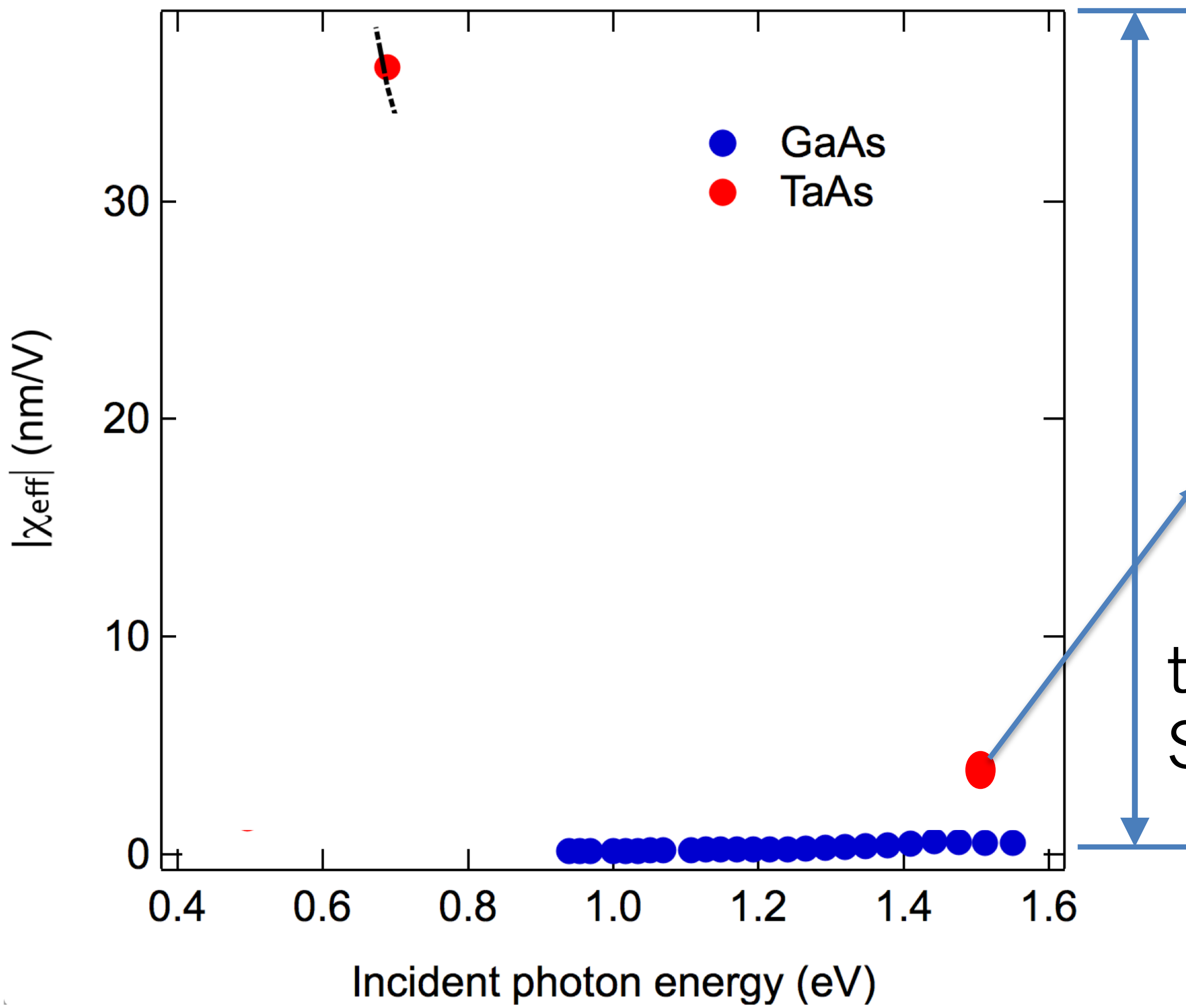
Material	$\lvert\chi_{ijk}\rvert$	$\lvert\chi\rvert$ (pm/V)	Fundamental wavelength (nm)
TaAs	χ_{zzz}	7200 (±1100)	800
GaAs	χ_{xyz}	700*	810
ZnTe	χ_{xyz}	500, 900*	800, 700
$BaTiO_3$	χ_{zzz}	30	900
$BiFeO_3$	χ_{zzz}	30-40	1550, 800
$LiNbO_3$	χ_{zzz}	50	852
$BiFeO_3$	χ_{zzz}	260*	500
$BaTiO_3$	χ_{zzz}	200*	170
$PbTiO_3$	χ_{zzz}	400*	150

Wu, *et al*. ***arXiv:1609.04894***
Nat. Phys. 13, 350(2017)

Spectroscopy of SHG response in range 0.4 eV – 1.6 eV



For fundamental and SH electric field along polar axis



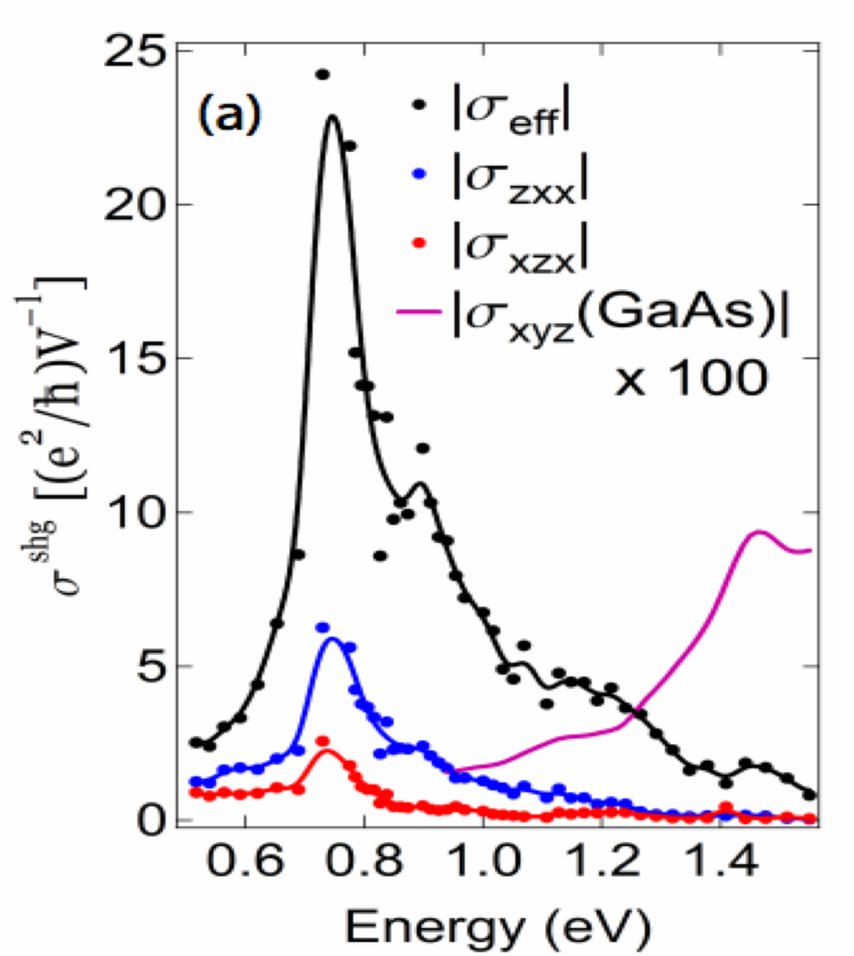
Wu, *et al*.
arXiv:1609.04894
Nat. Phys. (2017)

x 200^2=40,000

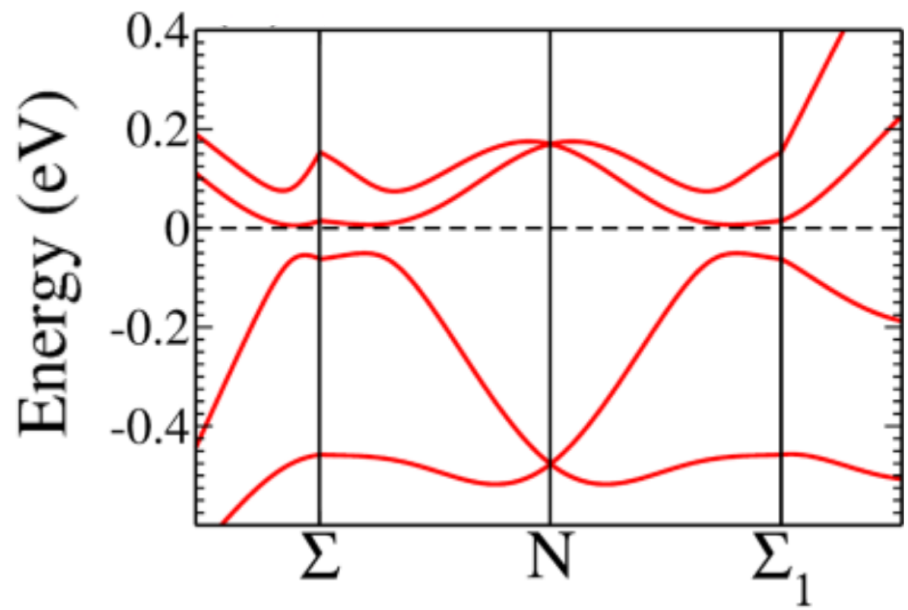
times more efficient SHG

arXiv:1804.06973, PRB (2018)

Resonance enhanced peak

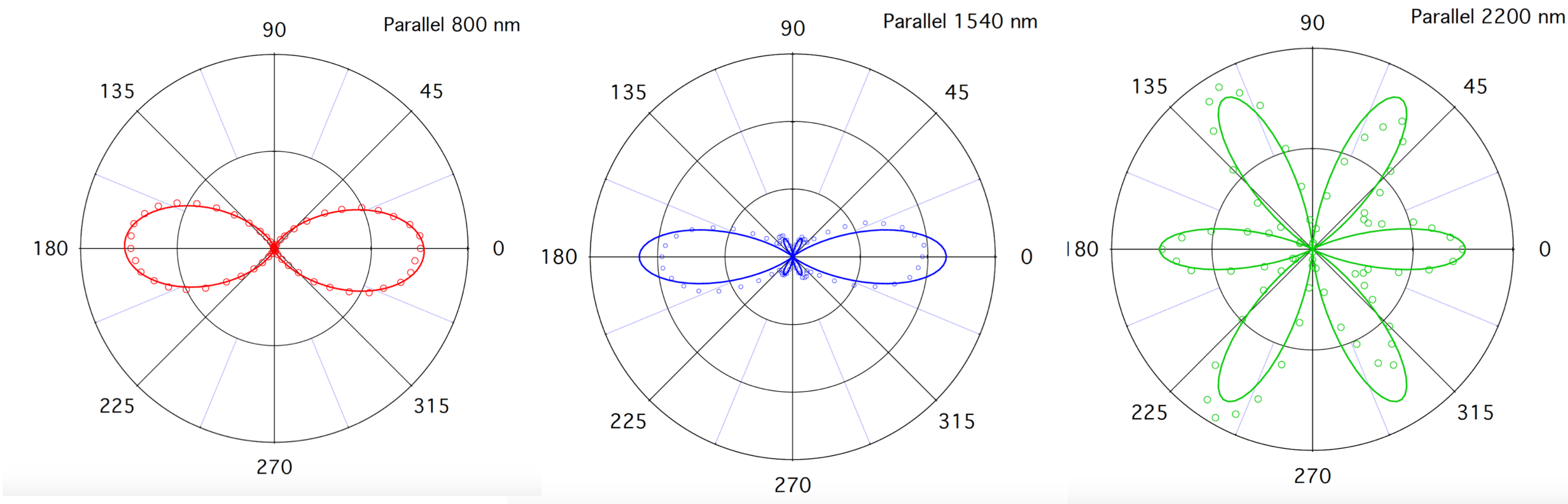


arXiv:1804.06973, PRB (2018)



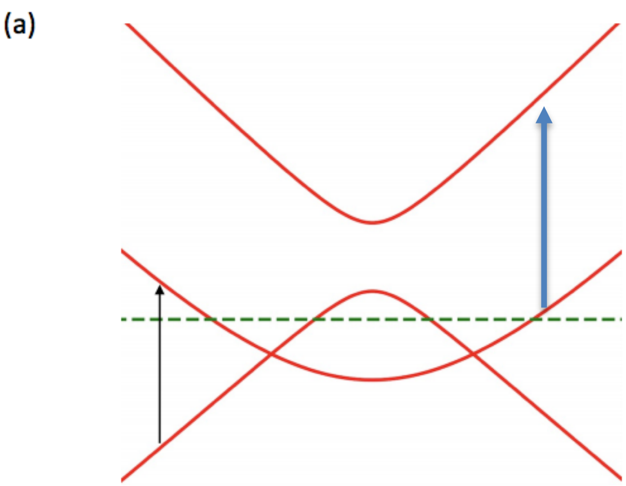
Weng *et al*., ***PRX*** (2015)
Huang, *et al*., ***Nat. Comm.*** (2015)
J. Buckeridge et al. ***PRB***, (2016)

Change of polar pattern

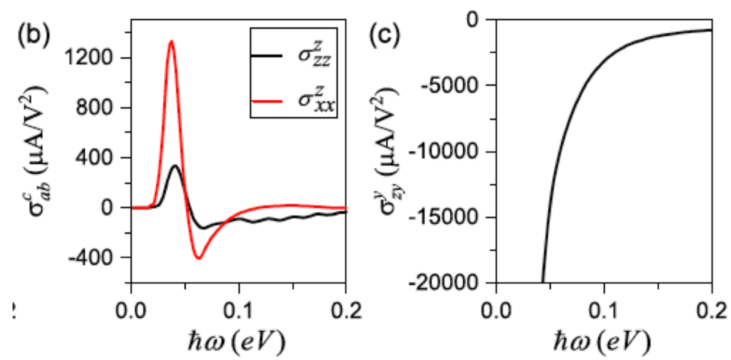


arXiv:1804.06973, PRB (2018)

Is Weyl physics related?



In addition, the calculated SH susceptibility χ^z_{zz} and the ratio of χ^x_{zx}/χ^z_{zz} are 6200 pm/V and 0.3 respectively, which are quite closed to the measured value 7200 pm/V and 0.031 at low temperature [45, 59].



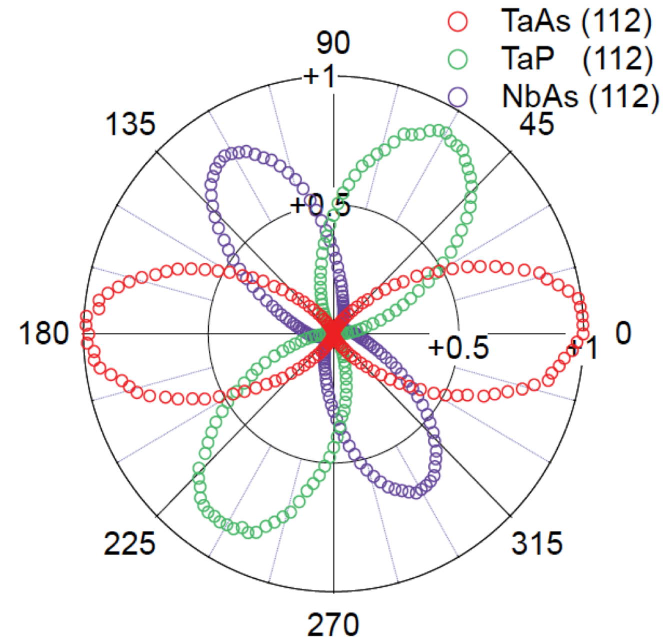
Zhang, ... Yan, Naogaosa ***arXiv.1803.00562, PRB (2018)***

Summary

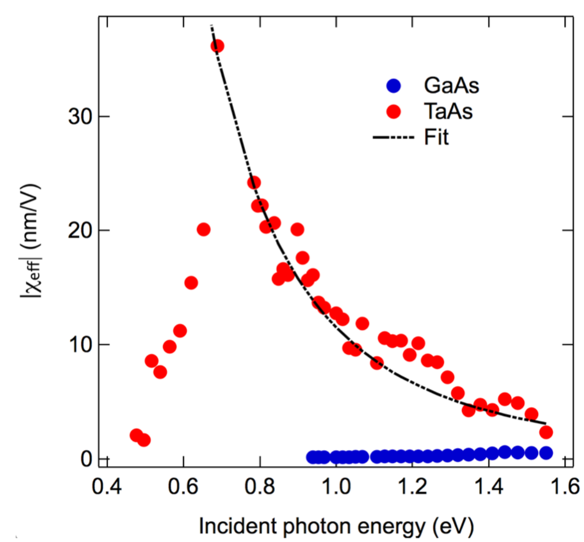
- **Discovery of the largest SHG in WSMs TaAs, TaP and NbAs**

- **A new perspective nonlinear optics in probing Berry connection/curvature**

Thanks !



Wu, *et al. arXiv:1609.04894*
***Nat. Phys. 13, 350* (2017)**



arXiv:1804.06973, PRB (2018)