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Structural transition and collapse of the gap in BiTeI under pressure

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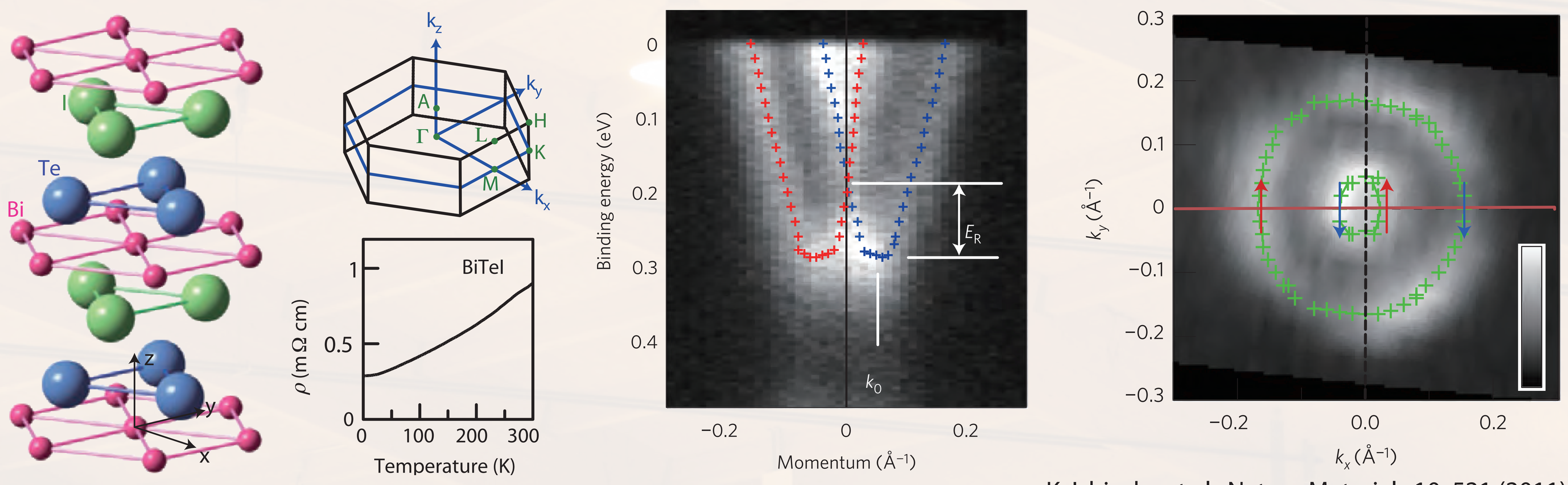
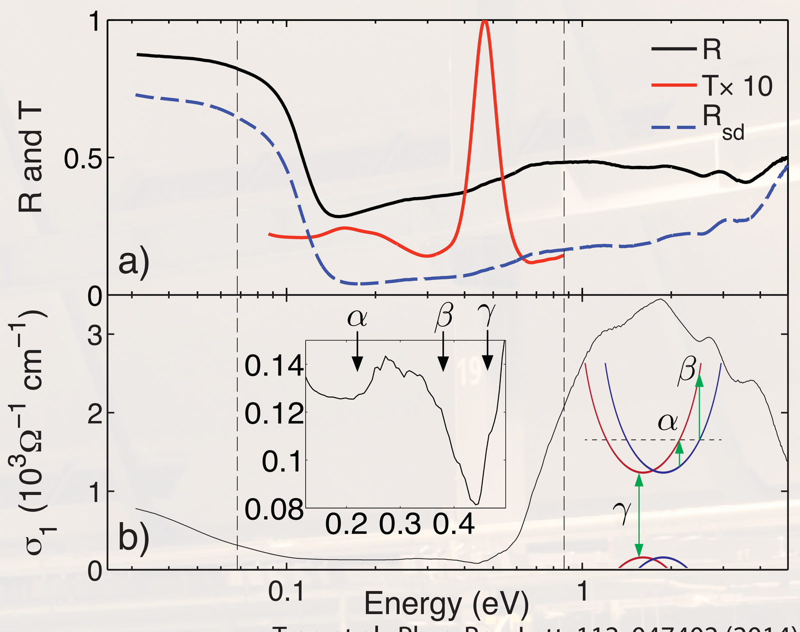
Motivations

- Samples grown by vapor flux method
- n-type semiconductor
- Low density of carriers n~10^{19} cm^{-3}
- Layered noncentrosymmetric structure P3m1
- Calculations & SR-ARPES shows Rashba spin splitting with large value of E_R ~ 0.1 eV

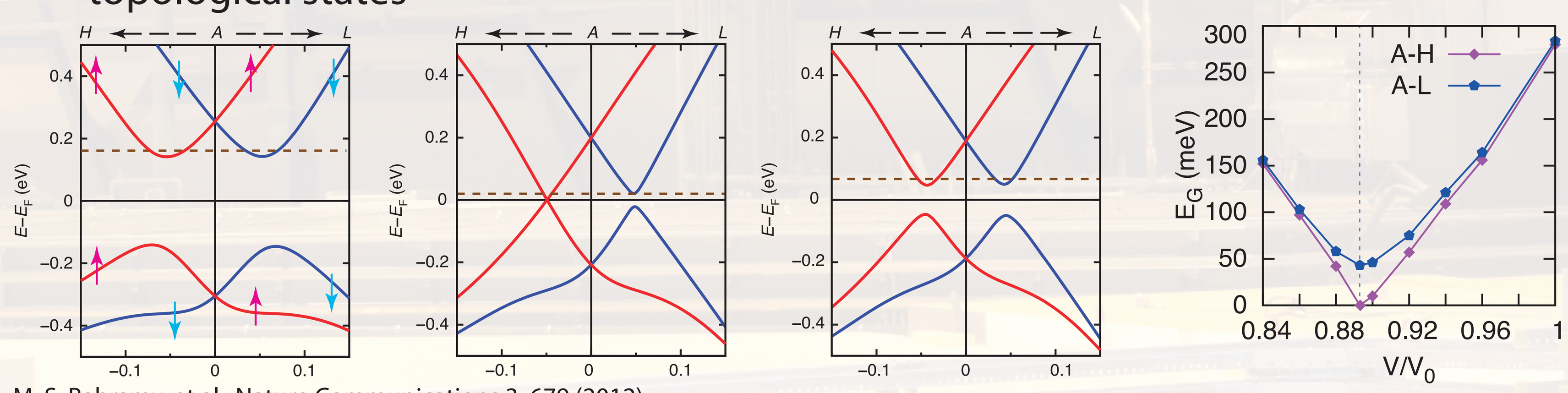


K. Ishizaka et al., Nature Materials 10, 521 (2011)

- Reflectivity *R* from FIR to VIS
- Variational dielectric function fit of *R* (Kramers-Kronig consistant) -> σ_1
- σ_1 contains a Drude contribution and is gaped until 0.4 eV with excitations between α and β of the RSS
- Infrared is a bulk technique, so RSS too
- DFT-GGA calculations predicted a P_c of 1.7-4.1 GPa with:
  - inversion of the band gap
  - quantum phase transition
  - unconventional metalicity
  - topological states



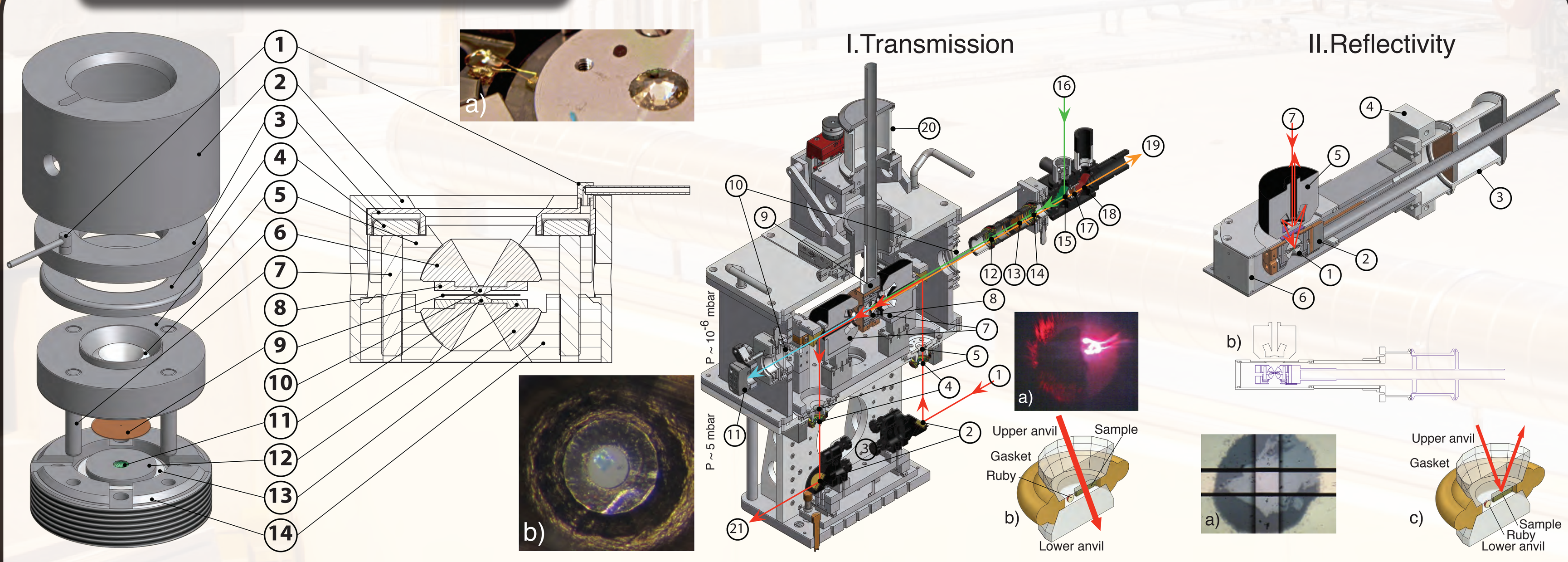
Tran et al., Phys. Rev. Lett. 112, 047402 (2014)



M. S. Bahramy, et al., Nature Communications 3, 679 (2012)

- Infrared spectroscopy under high pressure should allow to probe such transition

Experimental Setups



Left: BETSA MDAC 1. Membrane-to-capillary right angle adaptation piece, 2. Cover, 3. Membrane (stainless steel), 4. Interface disc, 5. Piston - upper rocker support (movable), 6. Upper rocker, 7. guide, 8. Upper diamond seat, 9. Gasket (CuBe), 10. Upper diamond, 11. Lower diamond, 12. Lower diamond seat, 13. Lower rocker, 14. Cylinder - lower rocker support (fixed). Insets: a) Cernox temperature sensor attached to the lower rocker and b) picture of the hole of the gasket with crystal sample, a ruby chip and liquid pressure media.

Right: Transmission and reflectivity setup in 3D cut view. Infrared is in red, laser excitation in green, ruby fluorescence in orange and visible light collected by camera in blue. **I.Transmission** 1. Incoming IR beam of the Bruker 66/v, 2. one inch Ø gold-plated steering mirrors on kinematic accuators, 3. Spectrometer's Focal Point and right Cassegrain rear FP, 4. Equivalent focal point with field-stop diaphrams, 5. IR-transparent windows separating upper and lower chambers, 6. 1 cm^2 square steering optics (mirror/beamsplitter/empty) on motorized linear stage, 7. 15x Cassegrain reflective objectives, 8. MDAC, 9. Cooldfinger tip, 10. Quartz windows, 11. CMOS camera, 12. Field-stop diaphragm, 13. Achromatic 50 mm doublet, 14. Aperture diaphragm, 15. Green dichroic mirror, 16. Green laser injection, 17. Achromatic 50 mm doublet, 18. Red dichroic mirror, 19. Fluoresece light collection, 20. Cold finger housing and 21. Outgoing IR beam. Insets: a) view of the gasket hole with synchrotron light spot next to crystal sample (reference spectrum); as seen with the camera in 11 and b) cut view of the sample environment with light passing tru the sample (sample spectrum). **II.Reflectivity** 1. Clamped MDAC, 2. Tip of the coldfinger, 3. ST-100 original housing, 4. Clamp for support/angle adjustment, 5. Hyperion 15x Reflective objective, 6. Low profile home-made housing and 7. Incoming/outgoing beam. Insets: a) view from the sample taken with the embedded camera of Hyperion with sample area delimited by field-stop blades (sample spectrum), b) the commons parts (in purple) shared with the transmission setup and c) sample environment in cut view.

M. K. Tran, et al., to be published

Data Analysis

Reflectivity data *R* at ambient pressure and R_{sd} at high pressure were analysed with the Kramers-Kroning consistant variational dielectric function technique (VDF*) of RefFIT to extract the high pressure optical conductivity σ_1 with the following workflow:

DL + KK VDF fit

$\hat{\epsilon}(\omega, P_0)$ as such

$$R(\omega, P_0) = \left|\frac{1-\sqrt{\hat{\epsilon}}}{1+\sqrt{\hat{\epsilon}}}\right|^2 \rightarrow \sigma_1(\omega, P_0)$$

starting point

→ DL + KK VDF fit

$\hat{\epsilon}'(\omega, P_0)$ as such

$$R(\omega, P_0) \approx \left|\frac{1-\sqrt{\hat{\epsilon}'}}{1+\sqrt{\hat{\epsilon}'}}\right|^2$$

&

$$R_{sd}(\omega, P_{i=1}) \approx C\left|\frac{\sqrt{\hat{\epsilon}_d}-\sqrt{\hat{\epsilon}'}}{\sqrt{\hat{\epsilon}_d}+\sqrt{\hat{\epsilon}'}}\right|^2$$

→ DL + KK VDF fit

$\hat{\epsilon}(\omega, P_{i=1})$ as such

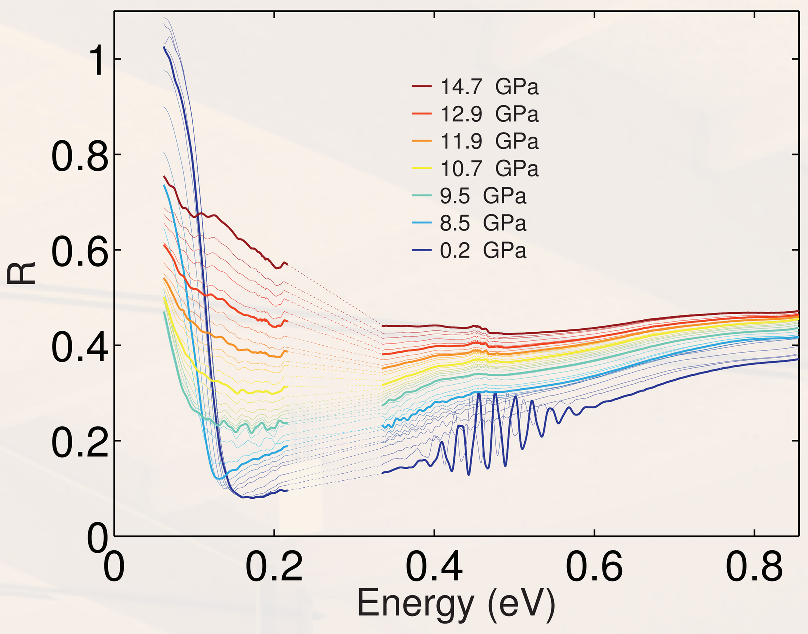
$$R_{sd}(\omega, P_{i=1}) = C\left|\frac{\sqrt{\hat{\epsilon}_d}-\sqrt{\hat{\epsilon}}}{\sqrt{\hat{\epsilon}_d}+\sqrt{\hat{\epsilon}}}\right|^2 \rightarrow \sigma_1(\omega, P_{i=1})$$

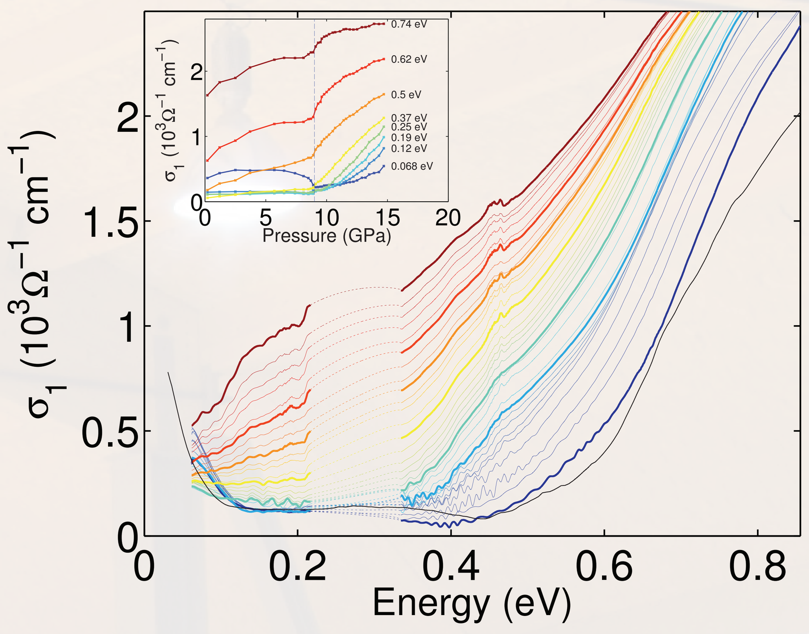
repeated step increasing index *i* & fixed *C*

C a scaling constant & $\hat{\epsilon}_d$ the dielectric function of diamond

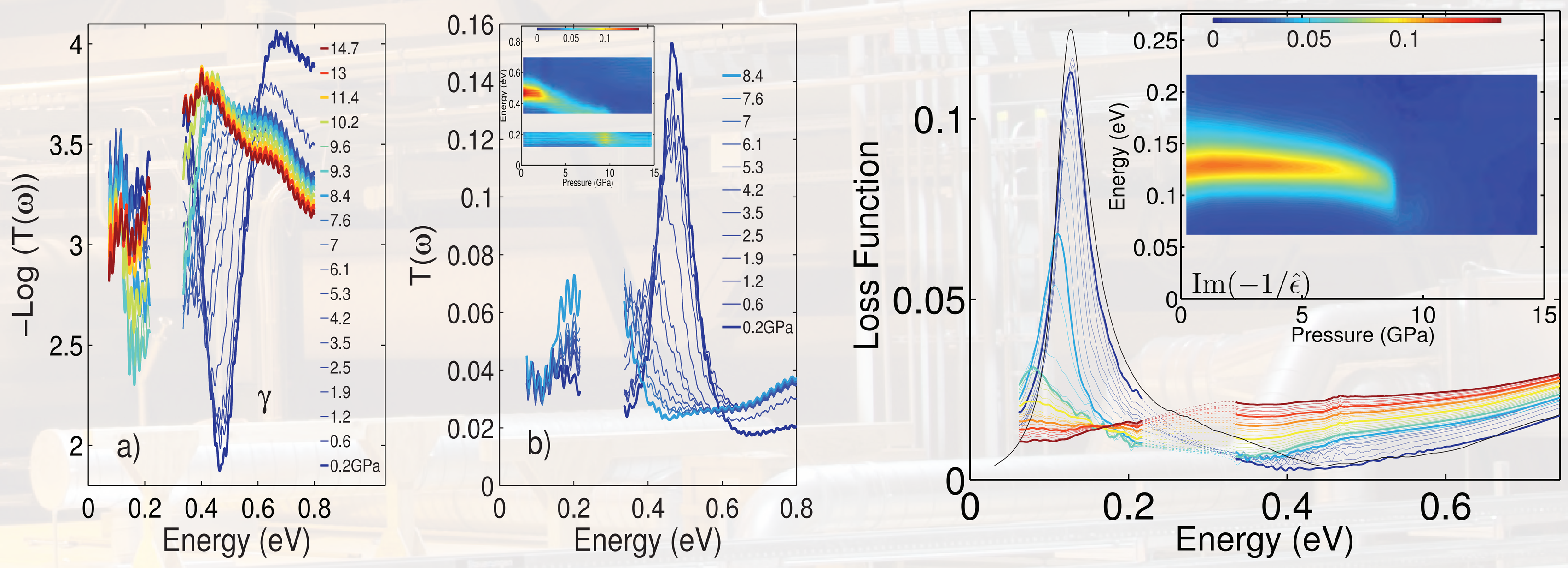
* A. B. Kuzmenko, Review of Scientific Instruments 76, 083108 (2005)

Results

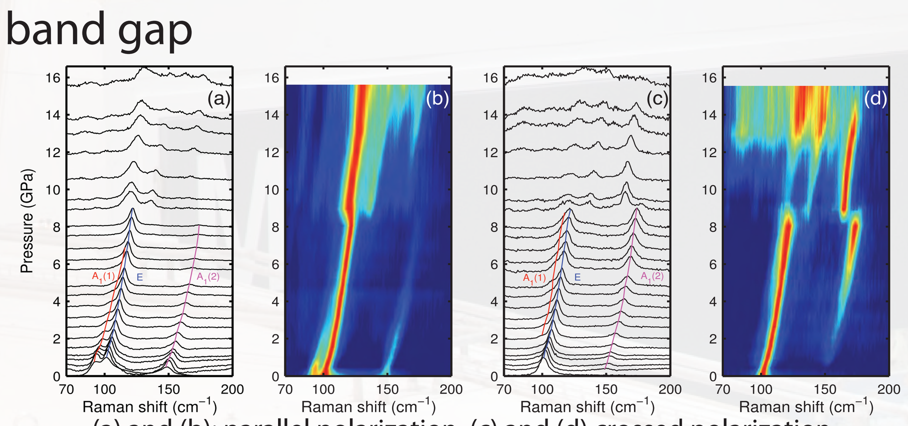




- Reflectivity
  - Reshift of plasma edge, P < 9 GPa
  - Increase (decrease) of R above (below) plasma edge
  - Vanishing of plasma edge, P > 9.5 GPa
- Conductivity
  - Redshift of absoption edge & disapearance of dip @ 0.45 eV for P < 8.5 GPa
  - Steep increase at 9 GPa (electronic transition)
  - Gapped states filled by 12 GPa and gap tends to very low energies
  - At 15 GPa, conductivity appears ungaped
  - Single peak (intraband plasmon) in Loss function disapears above 9 GPa

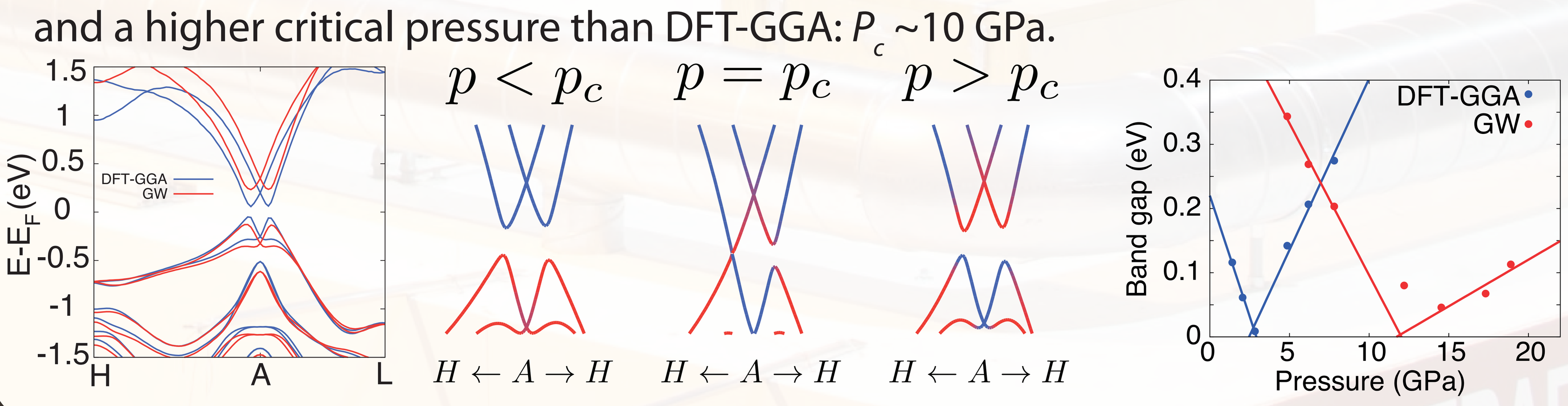


- Transmission shows
  - Single peak shifts to low energy between 3 and 9 Gpa above which it collapses
  - Phase transition & reduction of the band gap
- Polarized Raman indicates
  - Hardening of modes with pressure
  - Appearance of new modes at 9 GPa
  - Structural phase transition at 9 GPa



(a) and (b): parallel polarization. (c) and (d) crossed polarization.

- GW calculations indicate higher value for band gap (~0.4 eV - closer to experiment) and a higher critical pressure than DFT-GGA: P_c ~10 GPa.



Discussion

- Collapse of band gap with pressure, no re-opening is seen (hence no transition to a Topological Insulator phase)
- Structural transition at 9 GPa
- GW calculations set P_c ~12 GPa: prevents topological high pressure phase

- Identification of structure of high pressure phase
- Other study (X. Xi et al., Phys. Rev. Lett. 111, 155701 (2013)) concurs with the structural phase transition at ~8 GPa but claims the closing and reopening of the semiconducting gap through band inversion for P ~2-3 GPa
- Possible candidate for HP phase: $P6_3mc$ (doubling of unit cell perp. to layers & topologically trivial metallic state under pressure)

Tran et al., Phys. Rev. Lett. 112, 047402 (2014)