

Cd_3As_2 : a 3D analogue of graphene

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 Q. D. Gibson, R. J. Cava

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 Princeton University, USA

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 F. Teppe, W. Desrat

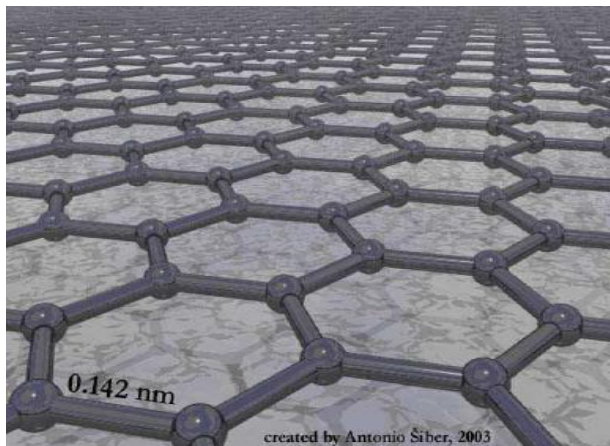
LNCMI, CNRS, Grenoble, France
 LCC, CNRS & Université Montpellier, France

Outline:

- Massless electrons in solid-state physics (reminder)
- 3D massless electrons in Cd_3As_2
- Conclusions

Graphene

2D



2D crystal made of carbon atoms organized in hexagonal lattice

Theoretically known over sixty years...

P. R. Wallace, Phys. Rev. 71, 622 (1947)

Isolated/fabricated in 2004/2005

K. S. Novoselov et al., Science 306, 666 (2004)

K. S. Novoselov et al., Nature 438, 197 (2005)

3D

Diamond



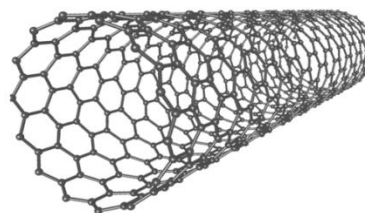
3D

Graphite



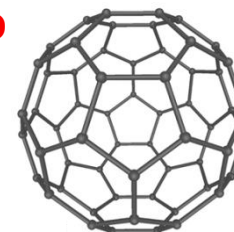
1D

Carbon nanotube



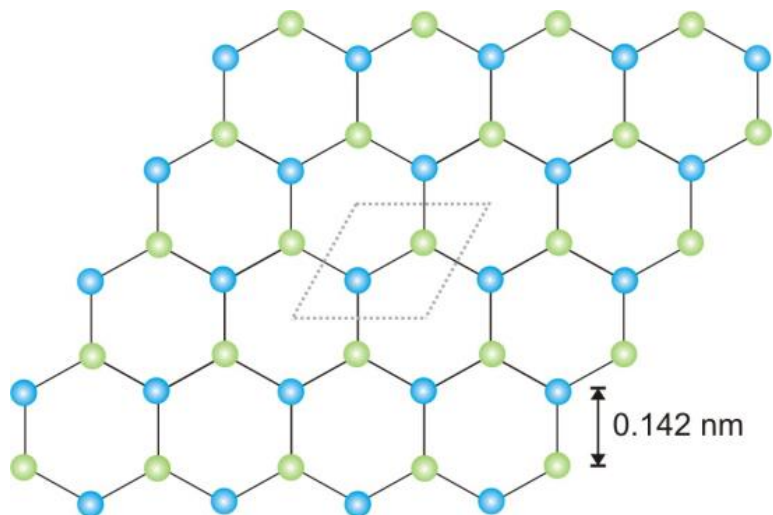
0D

Fulleren

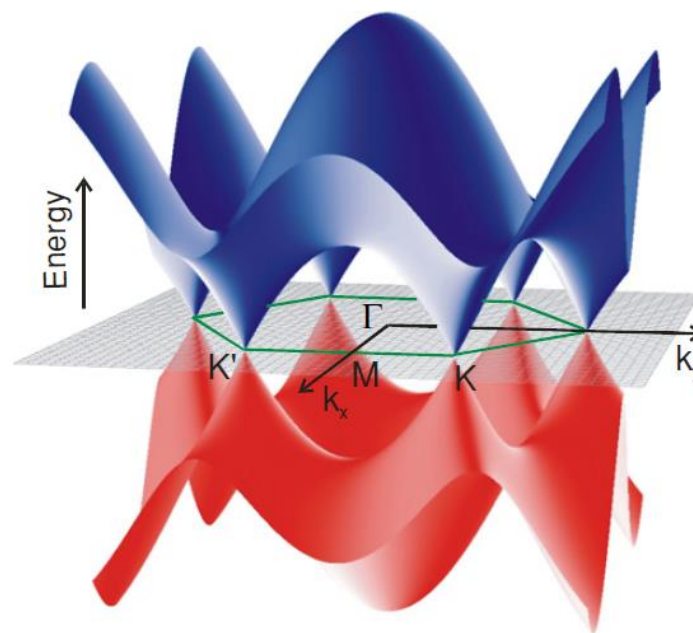


Electronic band structure of graphene

Crystal lattice:



Electronic bands:



Linearity of bands around K points:

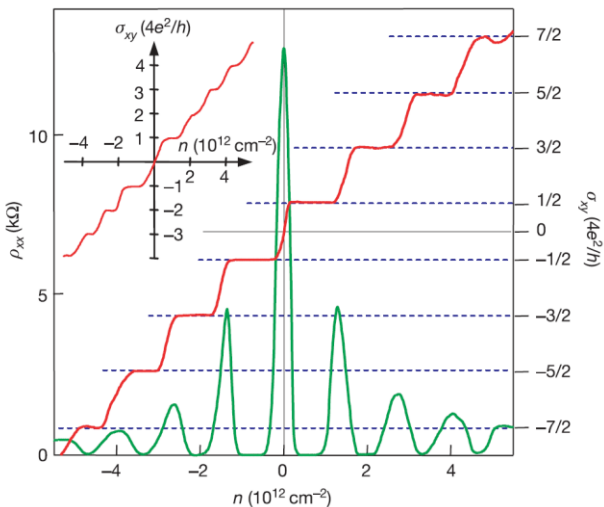
$$E(\mathbf{k}) \approx \pm \hbar v_F |\mathbf{k}| = \pm v_F |\mathbf{p}|$$

Electrons in graphene =
charged massless (relativistic) particles

$$E^2 = p^2 c^2 + \cancel{m_0^2 c^4}$$

Highlights of graphene physics

“Half-integer” and fractional QHE:



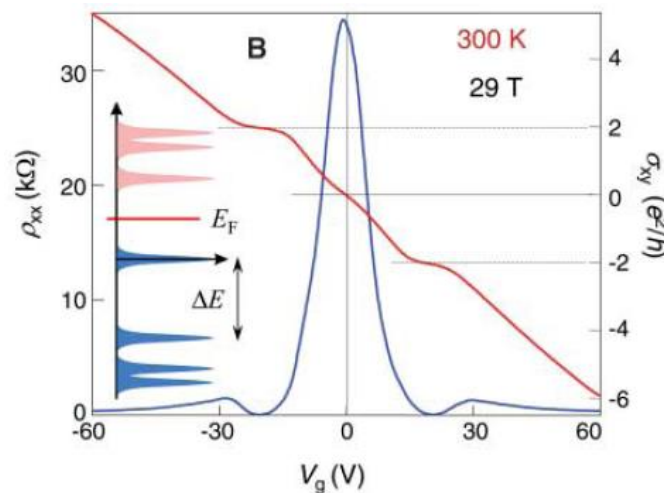
K. Novoselov et al., Nature 438, 197 (2005)

Y. Zhang et al., Nature 438, 201 (2005)

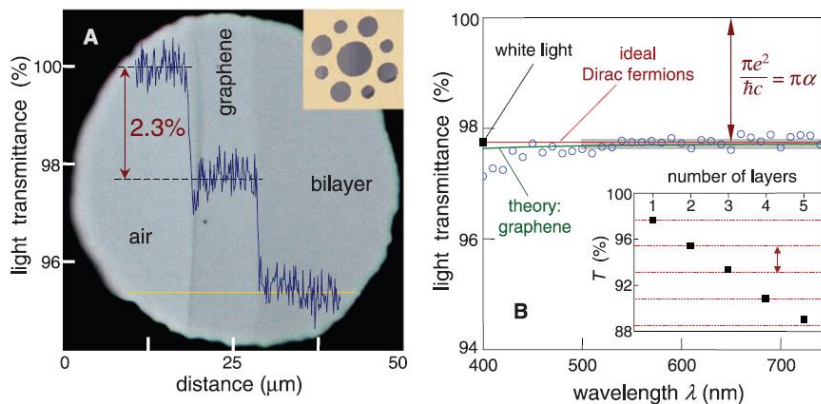
K. I. Bolotin et al., Nature 462, 196 (2009)

Room temperature QHE:

K. Novoselov et al., Science 315, 1379 (2007)



Universal dynamic conductivity:



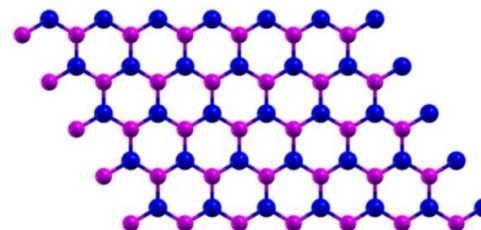
R. R. Nair et al., Science 320, 1308 (2008)

A. B. Kuzmenko et al., Phys. Rev. Lett. 100, 117401 (2008)

Other 2D Dirac-type materials

Silicene, germanene, phosphorene (?)

S. Cahangirov et al., Phys. Rev. Lett. 102, 236804 (2009)
 H. Liu et al., ACS Nano 8, 4033 (2014)

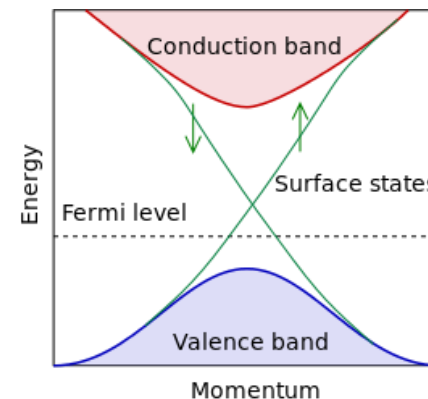
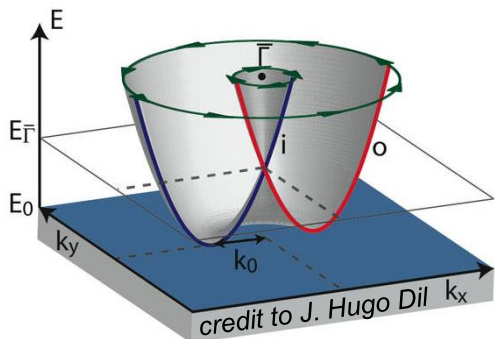


Artificial graphene, molecular graphene

C.-H. Park et al., PRL 101, 126804 (2008)
 M. Gibertini et al., Phys. Rev. B 79, 241406 (2009)
 L. Nádvořník et al., New J. Phys. 14, 053002 (2012)
 K. K. Gomes et al., Nature 483, 306 (2012)

Topological insulators (HgTe QWs, Bi₂Se₃, Bi₂Te₃, Bi_xSb_{1-x} etc.)

M. König et al., Science 318, 776 (2007)
 D. Hsieh et al., Nature 452, 970 (2008)
 H. Zhang et al., Nature Phys. 5, 438 (2009)



Semiconductors with a giant Rashba-type spin splitting (BiTeX, X = I, Cl or Br)

K. Ishizaka et al., Nature Materials 10, 521 (2011)
 S. Bordacs et al., Phys. Rev. Lett. 111, 166403 (2013)

Massless particles in 3D? Dirac and Weyl semimetals...

3D Dirac semimetals

(with spin-degenerate 3D conical bands)

Na_3Bi , Cd_3As_2

Z. K. Liu et al., *Science* 343, 864 (2014)

Z. K. Liu et al., *Nature Mater.* 13, 677 (2014)

S. Jeon et al., *Nature Mater.* 13, 851 (2014)

S. Borisenko et al., *Phys. Rev. Lett.* 113, 027603 (2014)

M. Neupane et al., *Nature Comm.* 5, 3786 (2014)

3D Weyl semimetals

(3D conical bands with no spin degeneracy)

TaAs, NbAs, TaP

B. Q. Lv et al., *Nature Phys.* 11, 724 (2015)

L. X. Yang et al., *Nature Phys.* 11, 728 (2015)

S.-Y. Xu et al., *Nature Phys.* 11, 748 (2015)

Dirac semimetal Cd_3As_2 = stable 3D analogue of graphene

nature
materials

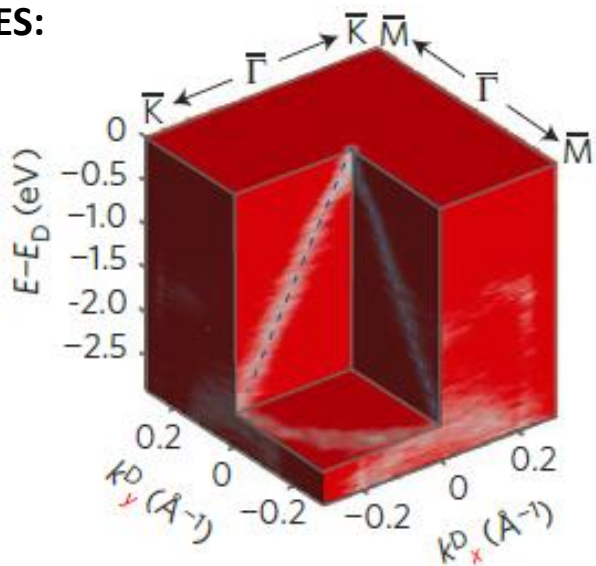
LETTERS

PUBLISHED ONLINE: 25 MAY 2014 | DOI: 10.1038/NMAT3990

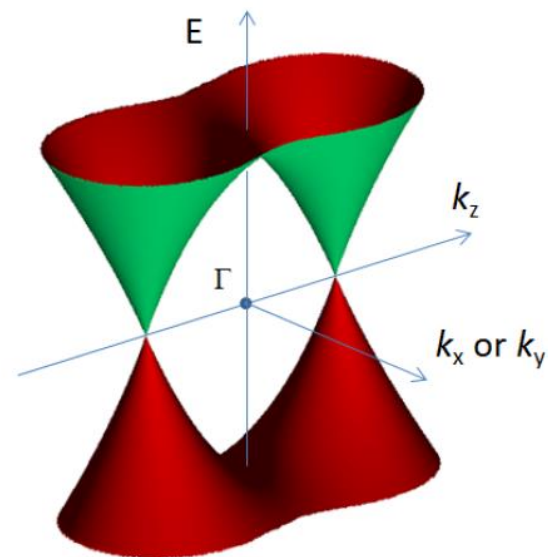
A stable three-dimensional topological Dirac semimetal Cd_3As_2

Z. K. Liu^{1†}, J. Jiang^{2,3†}, B. Zhou^{2,4†}, Z. J. Wang^{5†}, Y. Zhang^{1,4}, H. M. Weng⁵, D. Prabhakaran², S-K. Mo⁴, H. Peng², P. Dudin⁶, T. Kim⁶, M. Hoesch⁶, Z. Fang⁵, X. Dai⁵, Z. X. Shen¹, D. L. Feng³, Z. Hussain⁴ and Y. L. Chen^{1,2,4,6*}

ARPES:



3D Dirac semimetal Cd_3As_2



- Z. K. Liu et al., Nature Mater. 13, 677 (2014)
 S. Jeon et al., Nature Mater. 13, 851 (2014)
 S. Borisenko et al., Phys. Rev. Lett. 113, 027603 (2014)
 M. Neupane et al., Nature Comm. 5, 3786 (2014)

Dirac semimetal Cd_3As_2 = stable 3D analogue of graphene

nature
materials

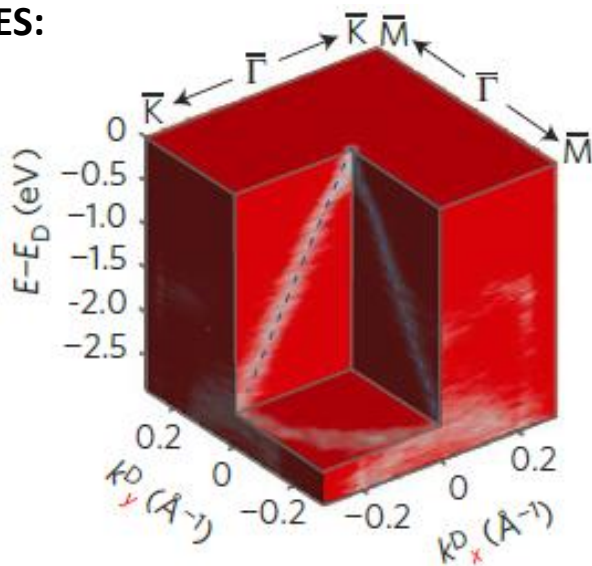
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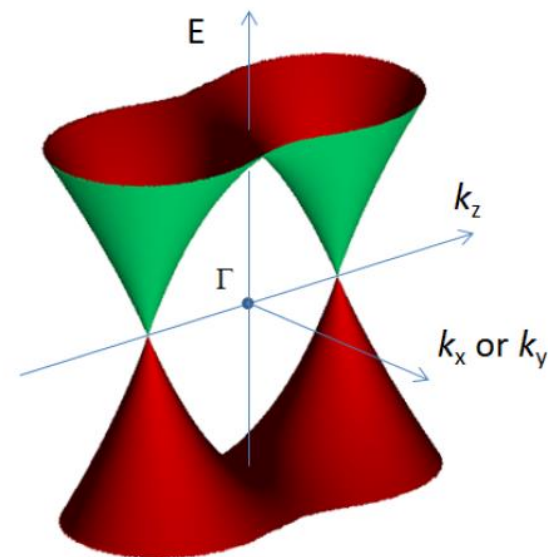
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ARPES:



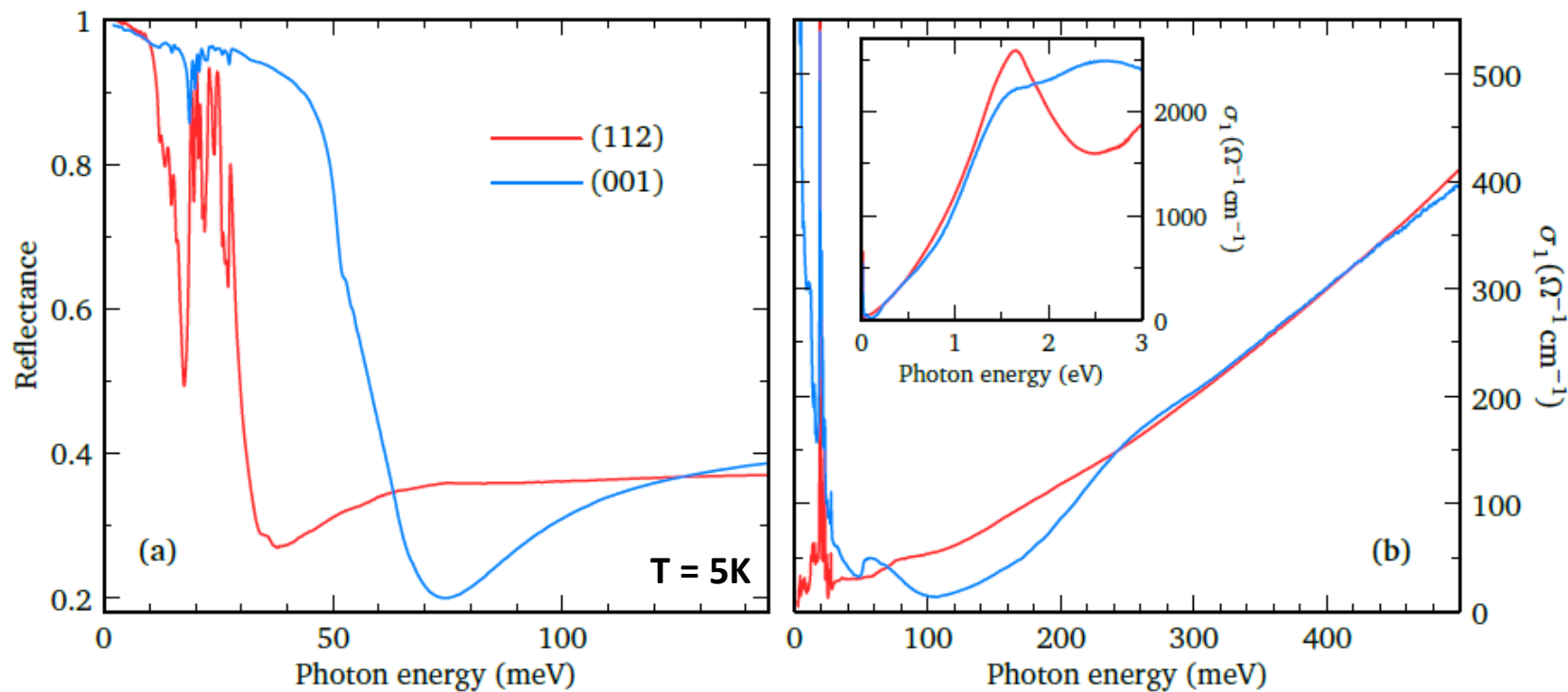
3D Dirac semimetal Cd_3As_2



...but really not in line with other studies:

- I. Rosenman, J. Phys. Chem. Solids 30, 1385 (1969)
- M. J. Aubin, et al., Phys. Rev. B 23, 3602 (1981)
- H. Schleijsen et al., Int. J. Infrared Milli. 5, 171 (1984)
- S. Jeon et al., Nature Mater. 13, 851 (2014)

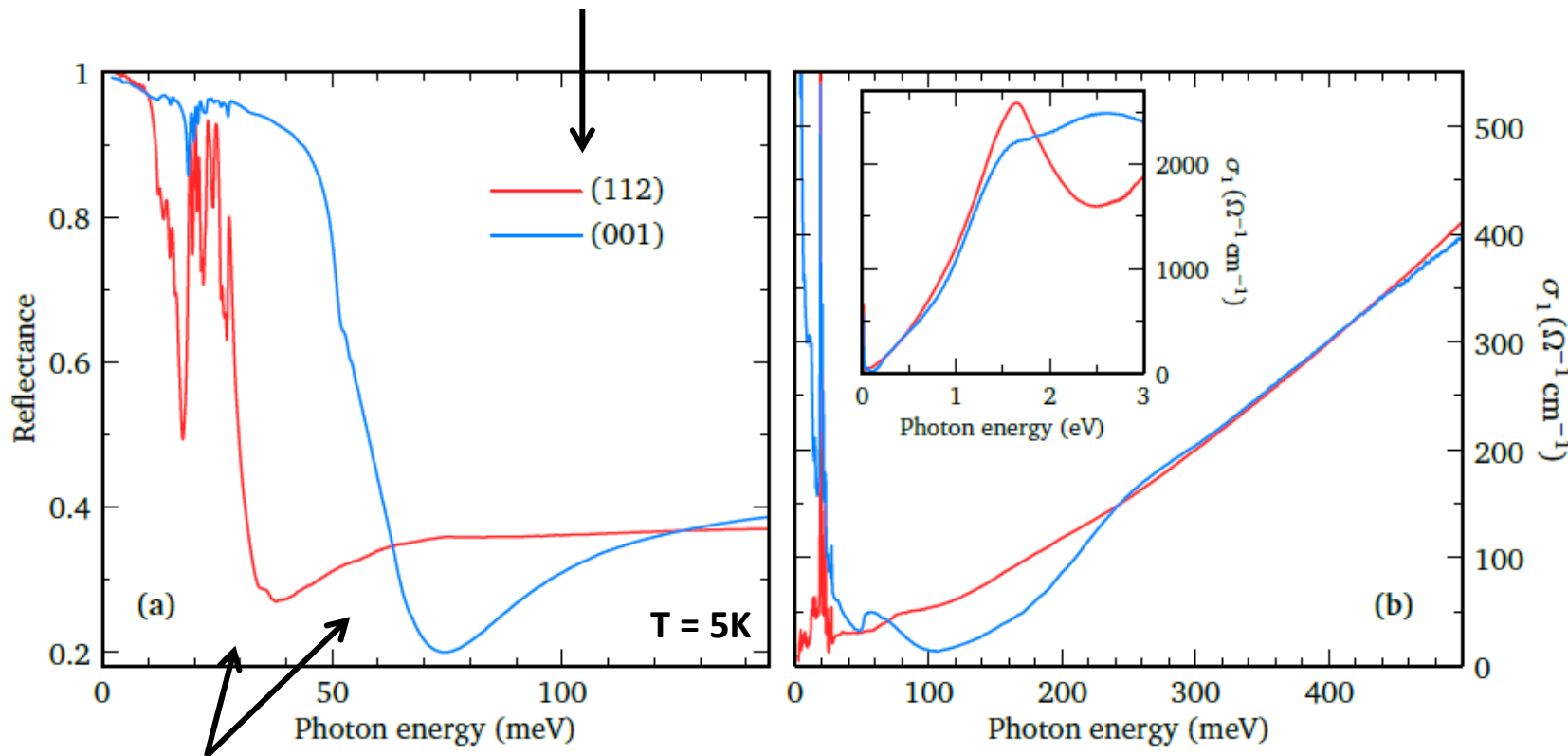
Cd_3As_2 – Infrared reflectance at $B=0$



...in collaboration with C. C. Homes (Brookhaven)

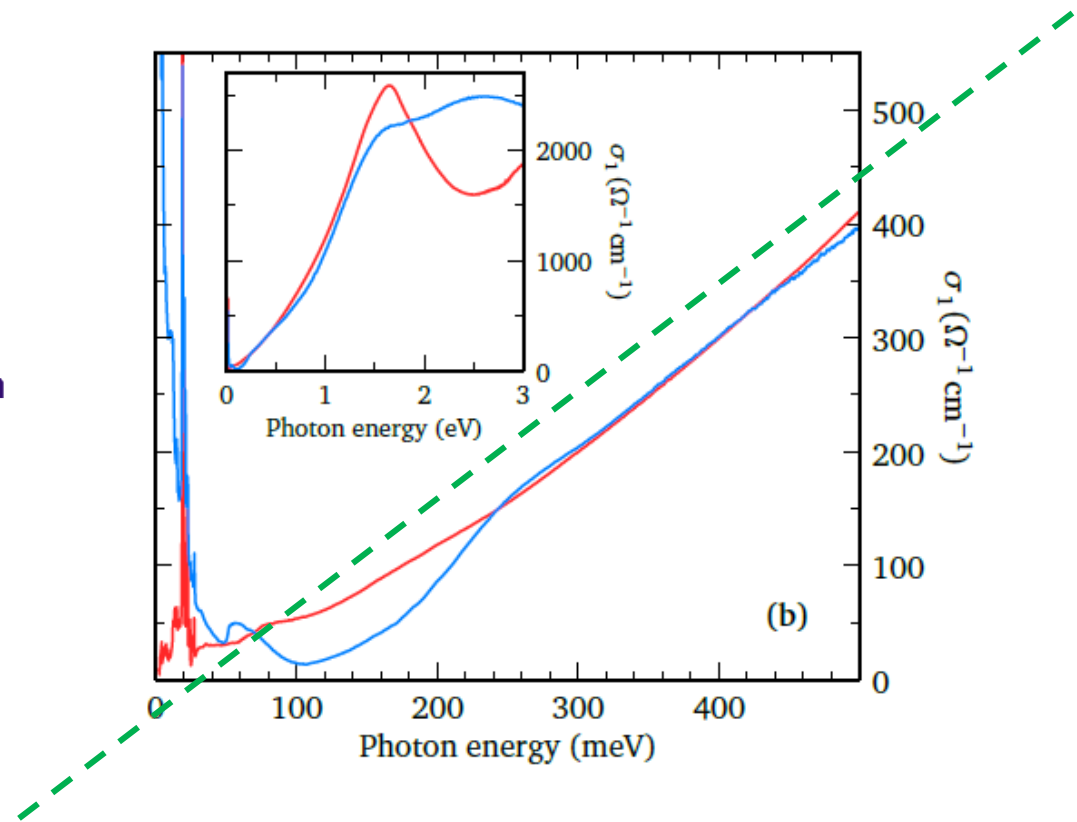
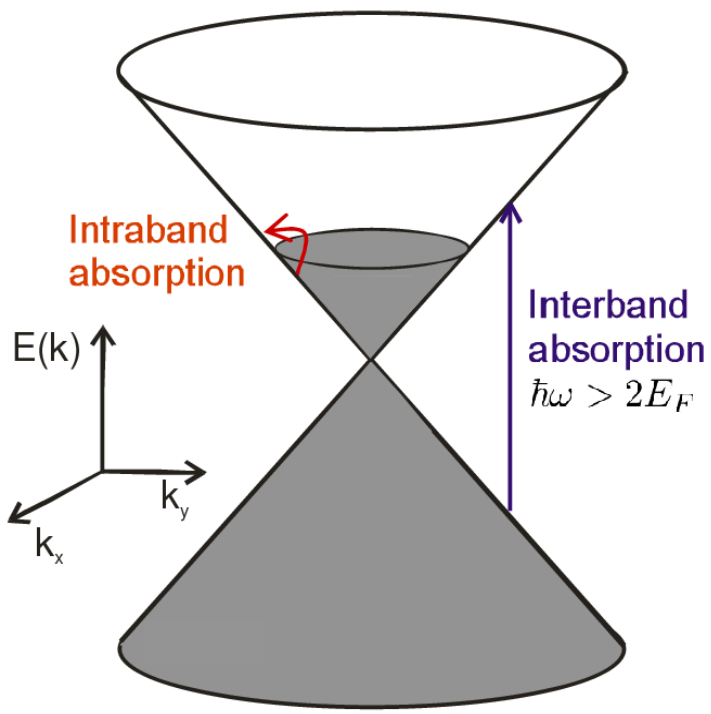
Cd_3As_2 – Infrared reflectance at $B=0$

Two different crystallographic orientations of **tetragonal** Cd_3As_2



Plasma edges
(different carrier densities)

Cd₃As₂ – Infrared reflectance at B=0



see also T. Timusk et al., Phys. Rev. B 87, 235121 (2013)
 M. Orlita et al., Nature Phys. 10, 233 (2014)

Cd₃As₂ – Infrared reflectance at B=0

Absorption of light in solids
(e.g., Fermi's golden rule):

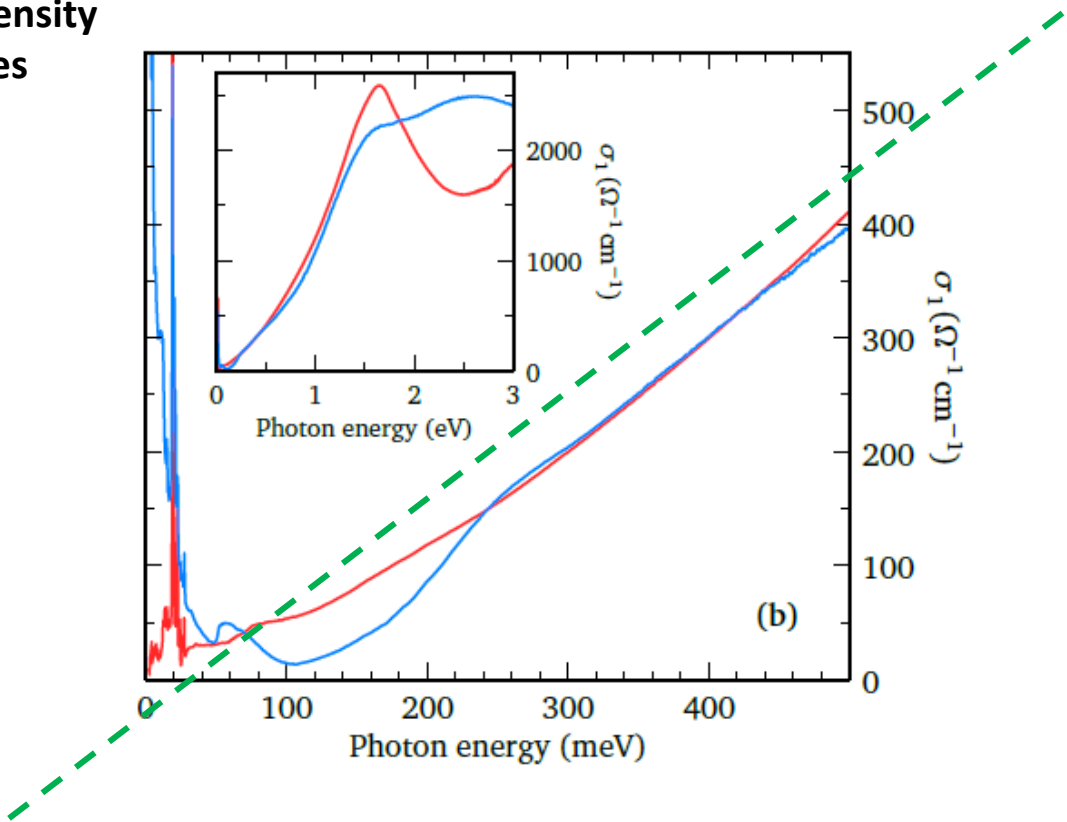
joint density
of states

$$\sigma_1(\omega) \propto \frac{D(\omega)}{\omega}$$

For conical bands in 3D:

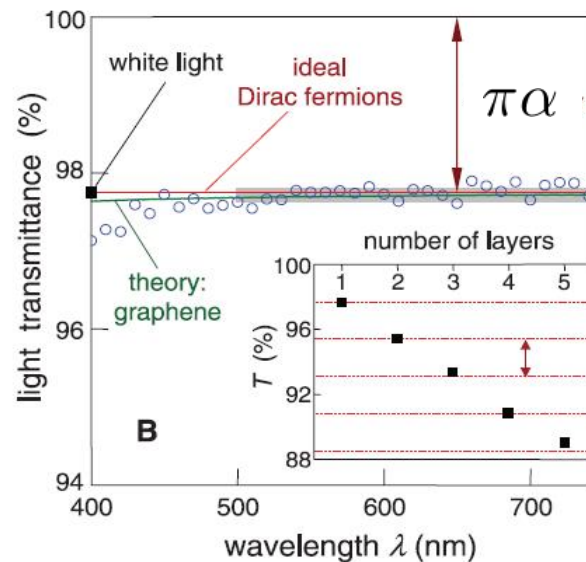
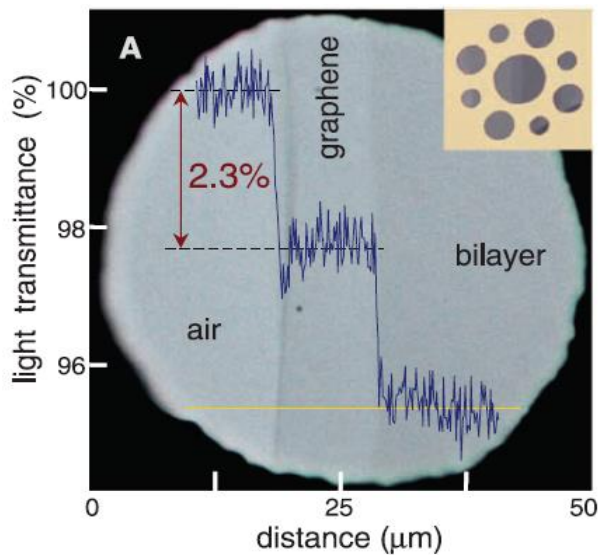
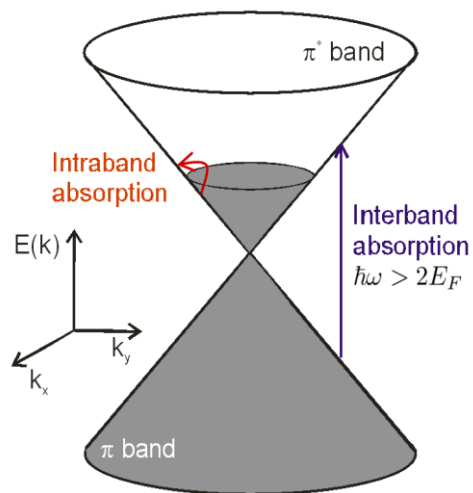
$$D(\omega) \propto \omega^2$$

Absorption coefficient (optical conductivity) linear in photon frequency!



see also T. Timusk et al., Phys. Rev. B 87, 235121 (2013)
M. Orlita et al., Nature Phys. 10, 233 (2014)

Absorption of light in graphene



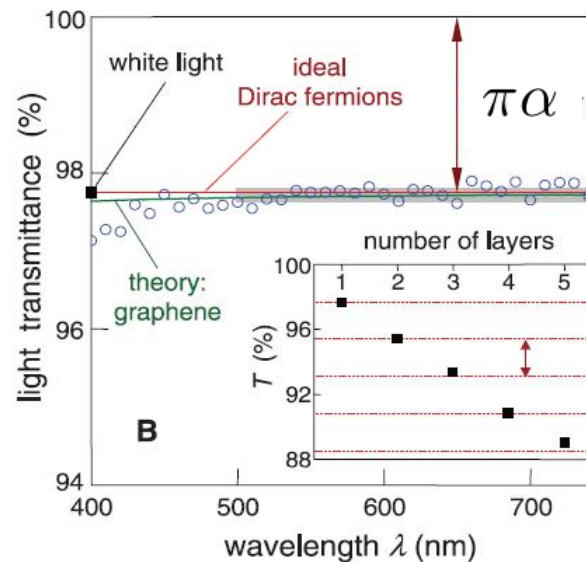
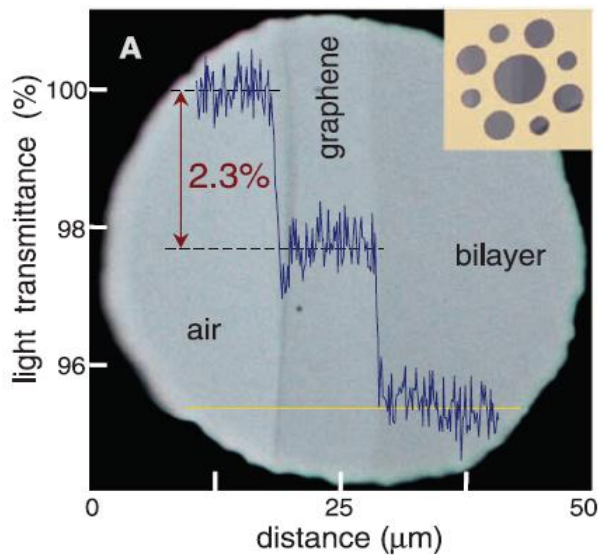
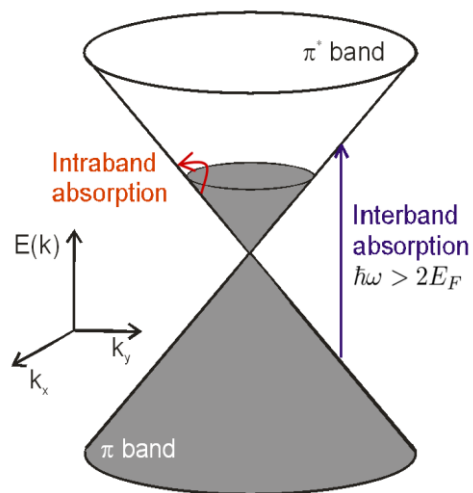
“Flat” absorption of light (2.3%) defined only by the fine structure constant:

$$\alpha = \frac{e^2}{4\pi\epsilon_0\hbar c} \doteq \frac{1}{137}$$

R. R. Nair et al., Science 320, 1308 (2008)

A. B. Kuzmenko et al., Phys. Rev. Lett. 100, 117401 (2008)

Absorption of light in graphene



Absorption of light in solids
(Fermi's golden rule):

$$\lambda(\omega) \propto \frac{D(\omega)}{\omega}$$

For conical bands in 2D:

$$D(\omega) \propto \omega$$

Dispersionless (interband) absorption of light...

Cd₃As₂ – Optical response at B=0

Absorption of light in solids
(e.g., Fermi's golden rule):

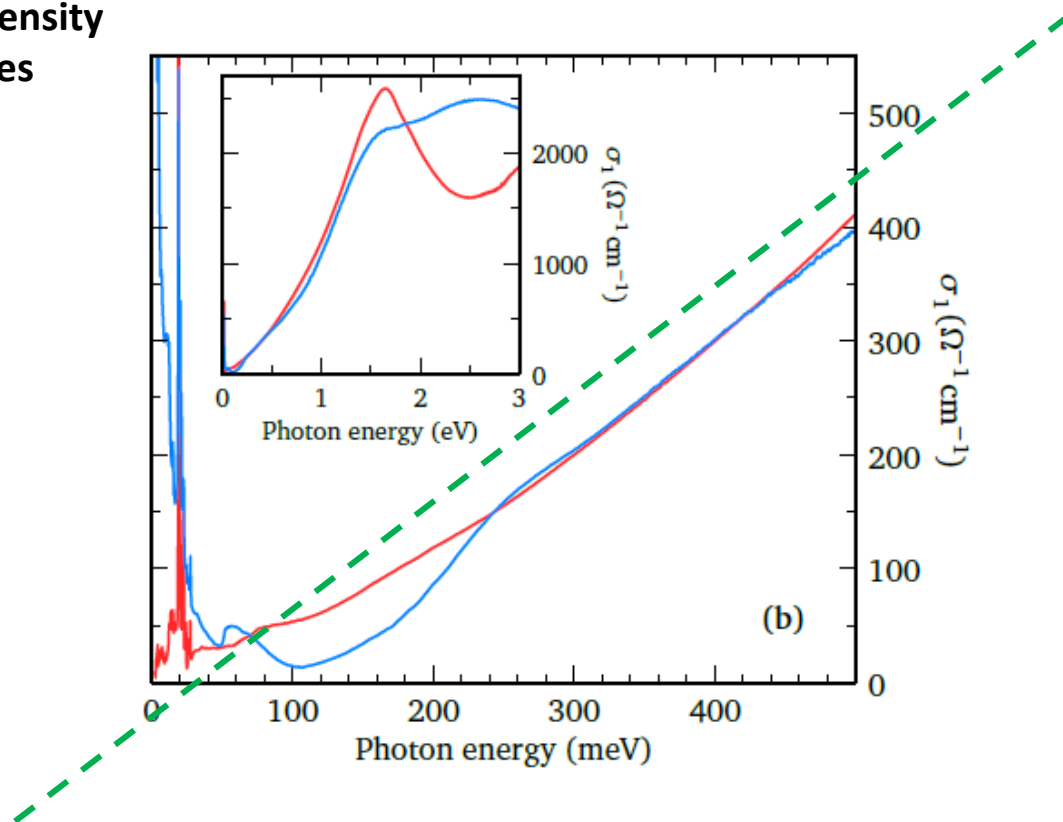
$$\sigma_1(\omega) \propto \frac{D(\omega)}{\omega}$$

joint density
of states

For conical bands in 3D:

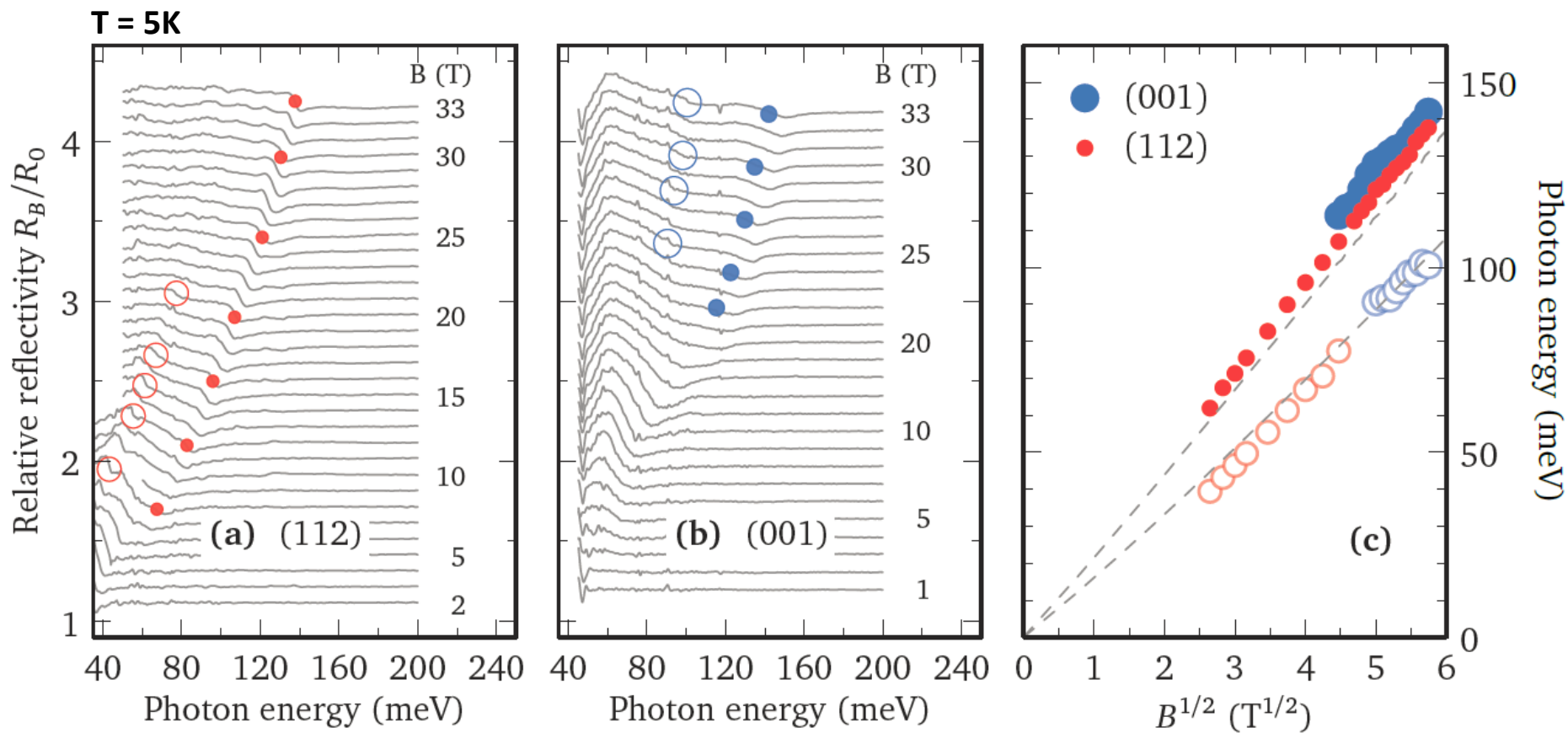
$$D(\omega) \propto \omega^2$$

**Absorption coefficient linear in
photon frequency!**



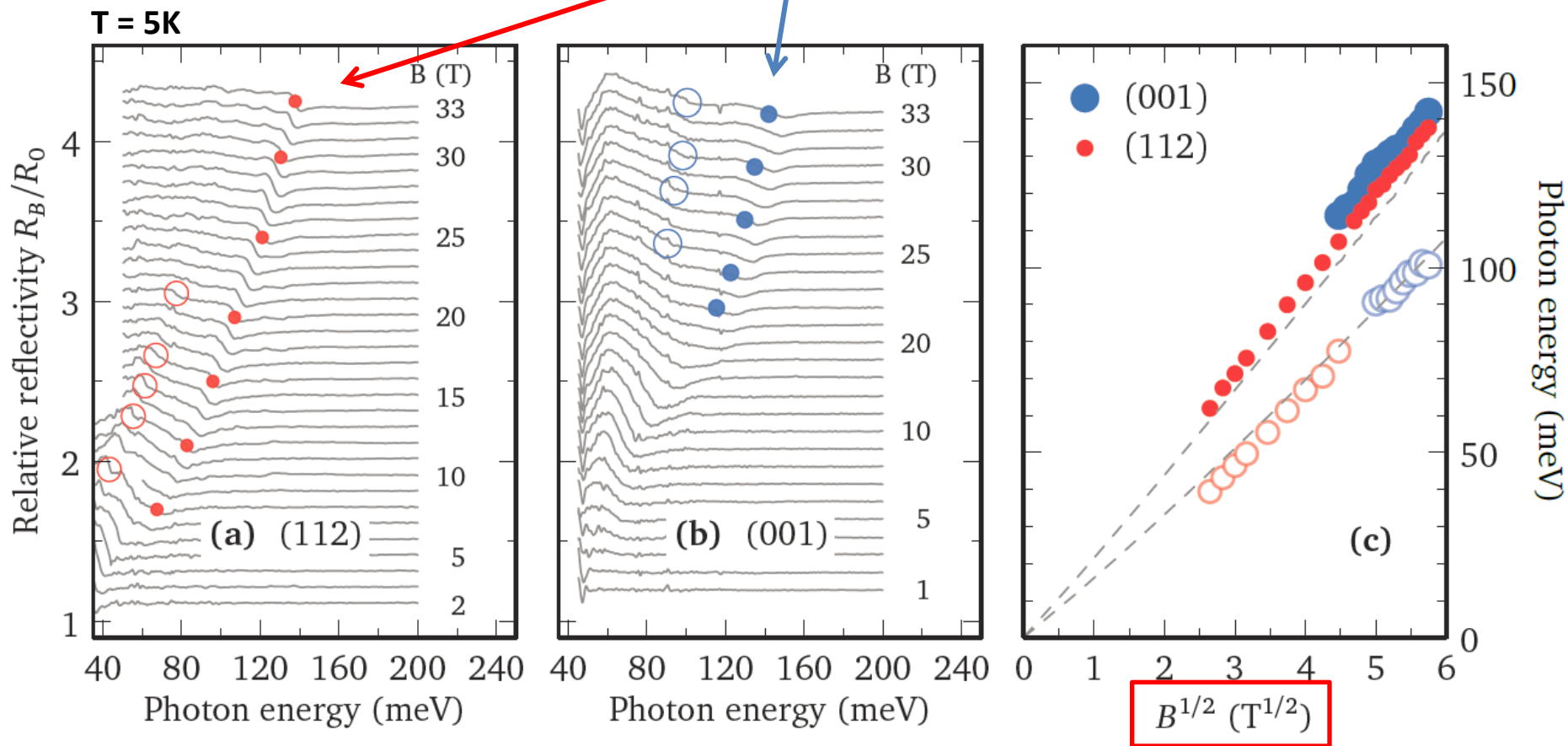
see also T. Timusk et al., Phys. Rev. B 87, 235121 (2013)
M. Orlita et al., Nature Phys. 10, 233 (2014)

Cd₃As₂ – High-field magneto-reflectivity



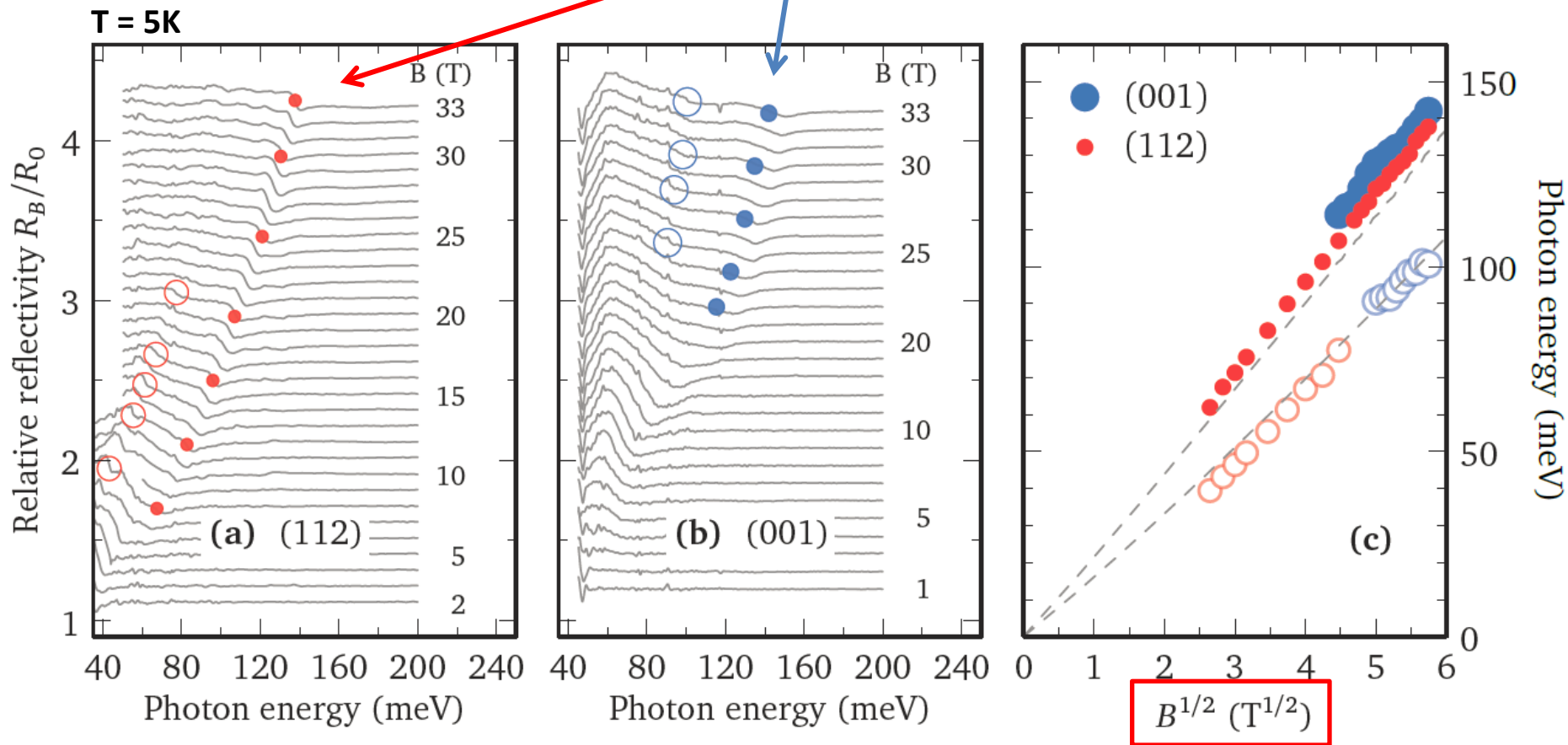
Cd₃As₂ – High-field magneto-reflectivity

Cyclotron resonance (CR) in the quantum limit



Cd₃As₂ – High-field magneto-reflectivity

Cyclotron resonance (CR) in the quantum limit

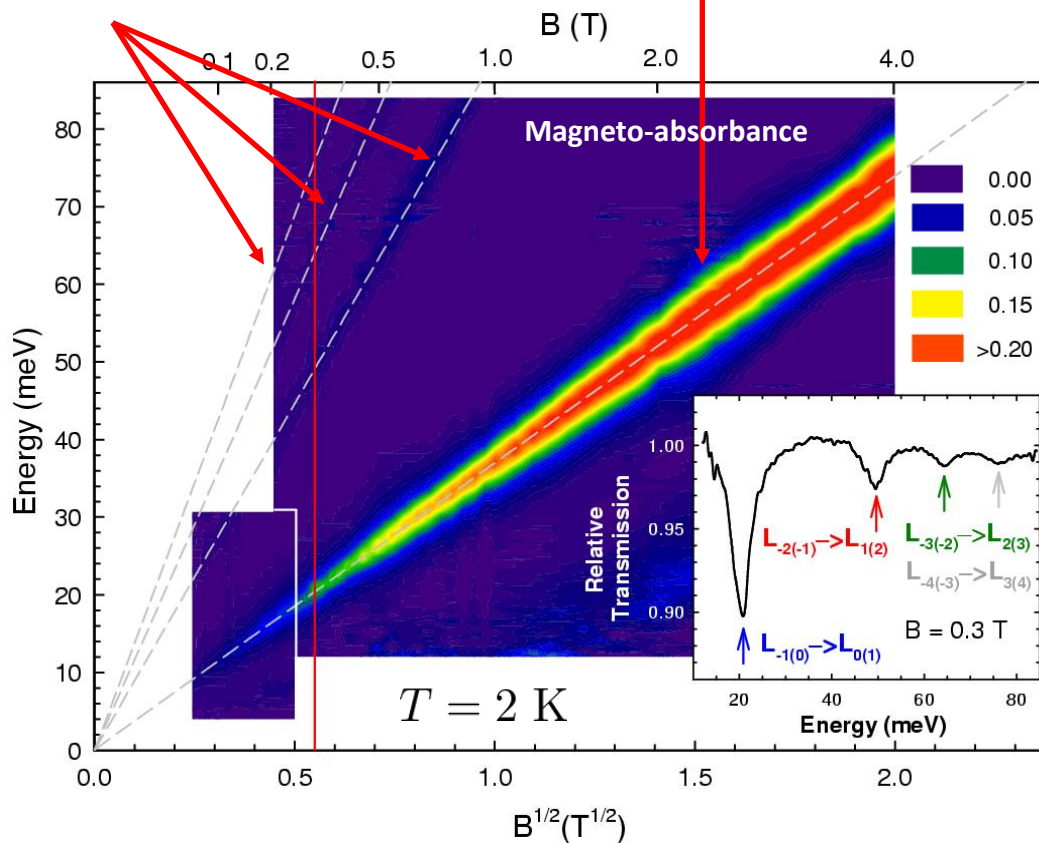


Magneto-optical response linear in \sqrt{B} = typical signature of massless particles

Magneto-transmission of (multilayer epitaxial) graphene

Interband inter-Landau level transitions

Cyclotron resonance



Energy spectrum:

$$E_n = \pm v \sqrt{2e\hbar B |n|}$$

Velocity:

$$v = 1.02 \times 10^6 \text{ m/s}$$

Selection rules:

$$|n| \rightarrow |n| \pm 1$$

Landau levels in 2D

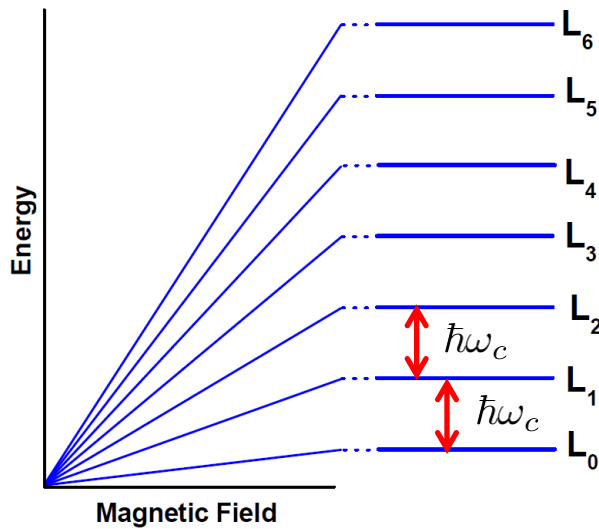
Parabolic band:

$$E = \frac{p^2}{2m}$$

Landau level spectrum:

$$E_n = \hbar\omega_c(n + 1/2)$$

$$\omega_c = eB/m$$

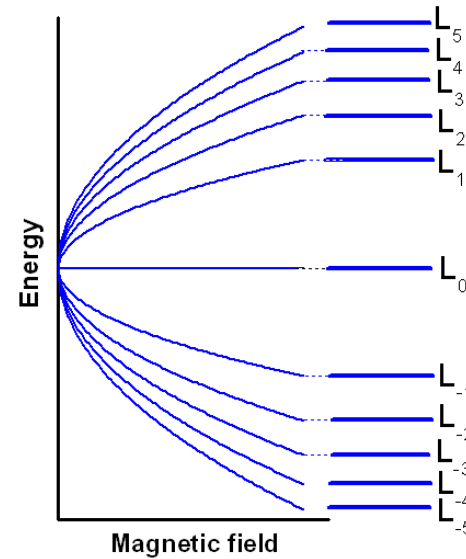


Conical band:

$$E = \pm v|p|$$

Landau level spectrum:

$$E_n = \pm v\sqrt{2e\hbar Bn}$$

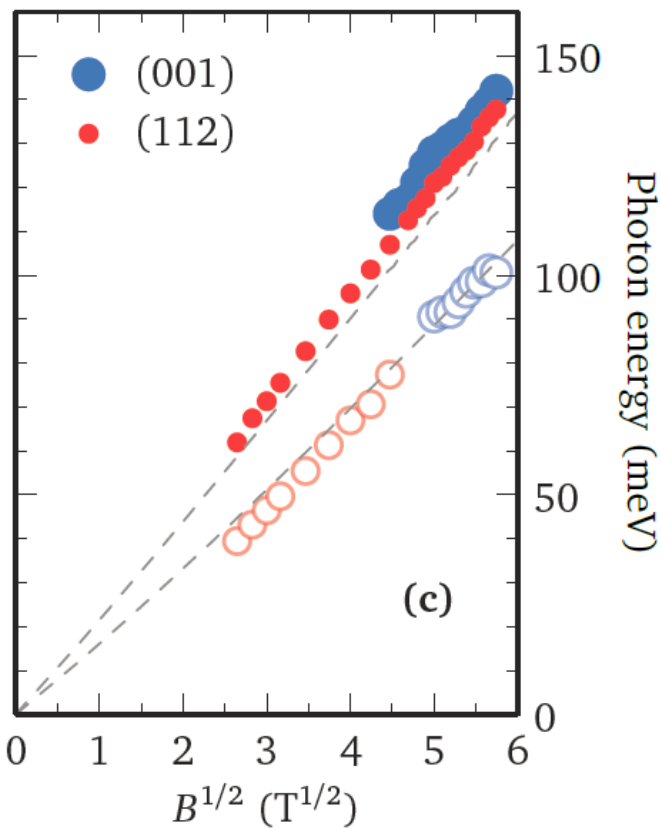
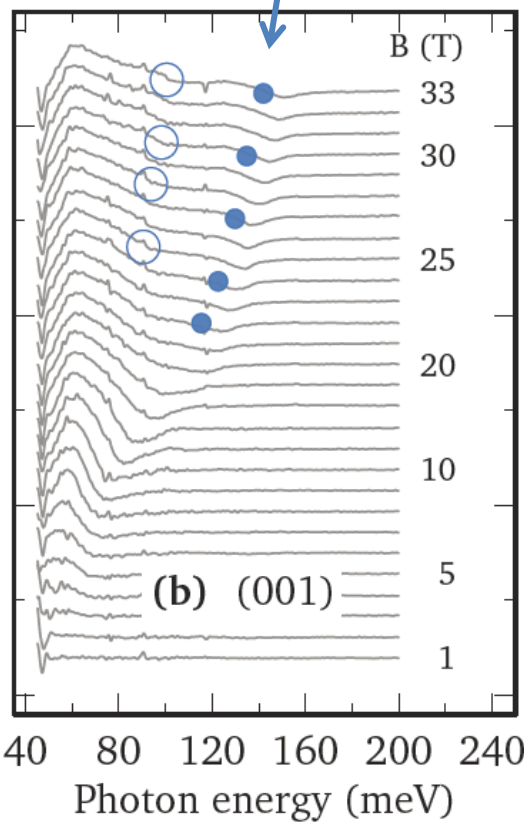
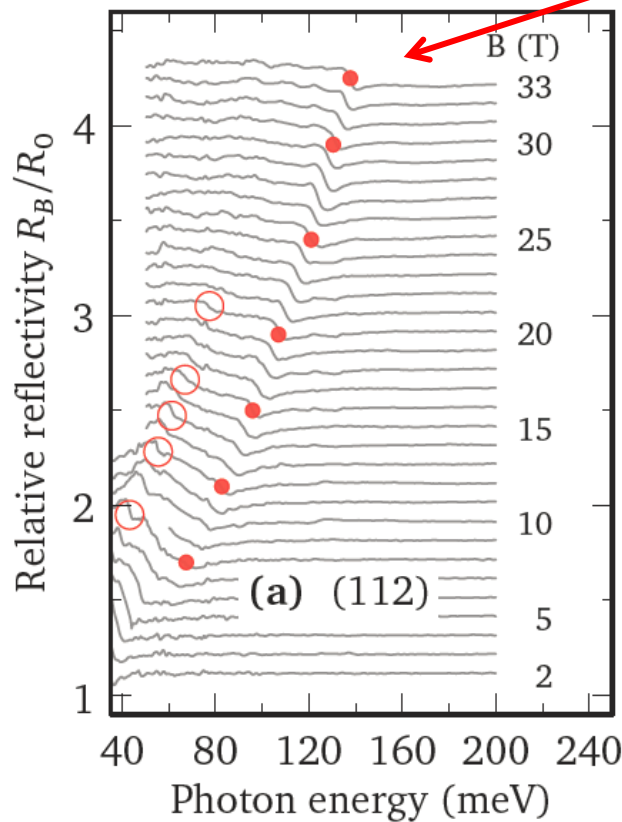


Electron LLs

Hole LLs

Cd₃As₂ – High-field magneto-reflectivity

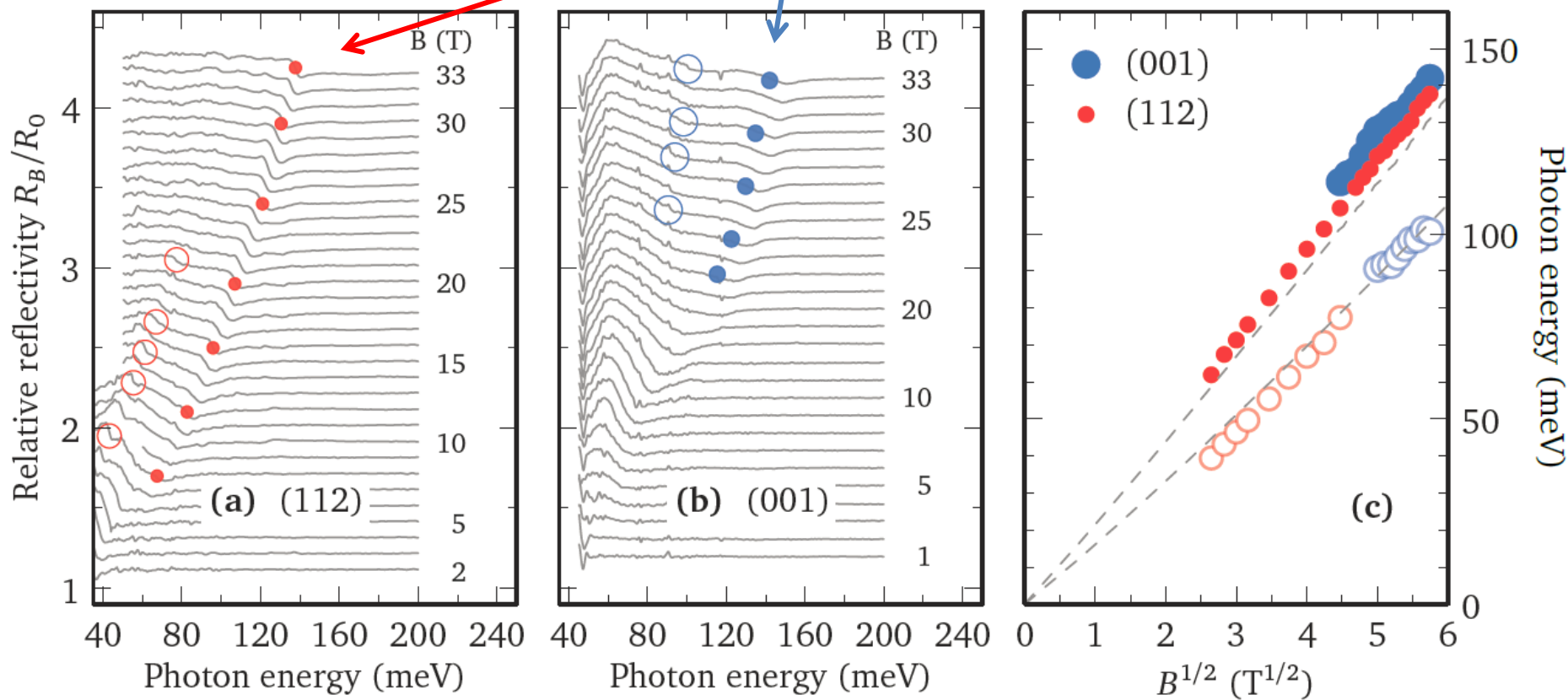
Cyclotron resonance (CR) in the quantum limit



Magneto-optical response linear in \sqrt{B} = typical signature of massless particles

Cd₃As₂ – High-field magneto-reflectivity

Cyclotron resonance (CR) in the quantum limit

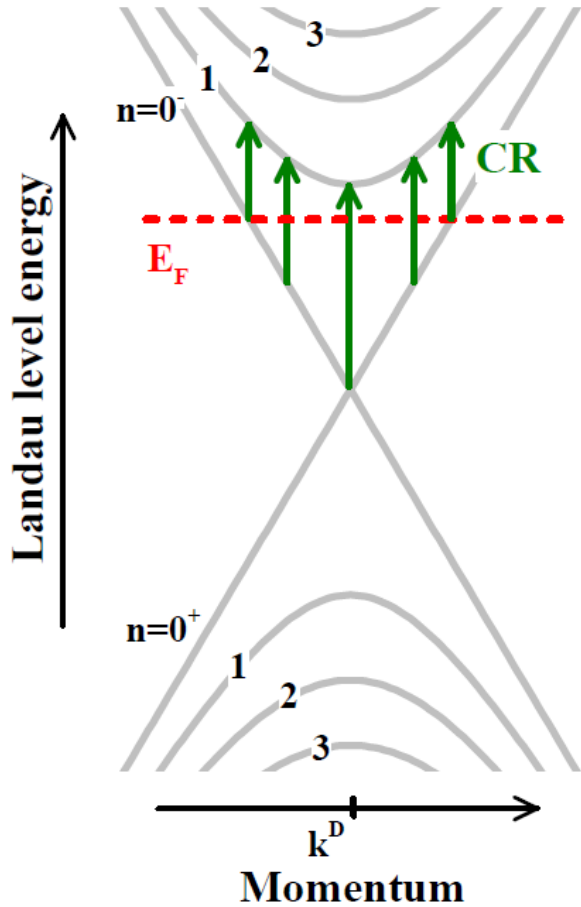


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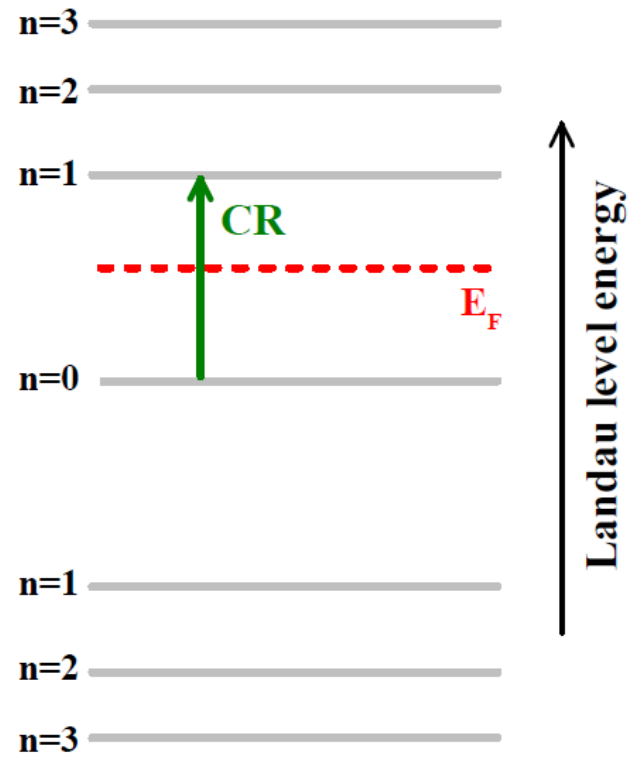
...massless yes, but not 3D Dirac

Dirac electrons – Landau level spectrum

3D Dirac electrons



2D Dirac electrons

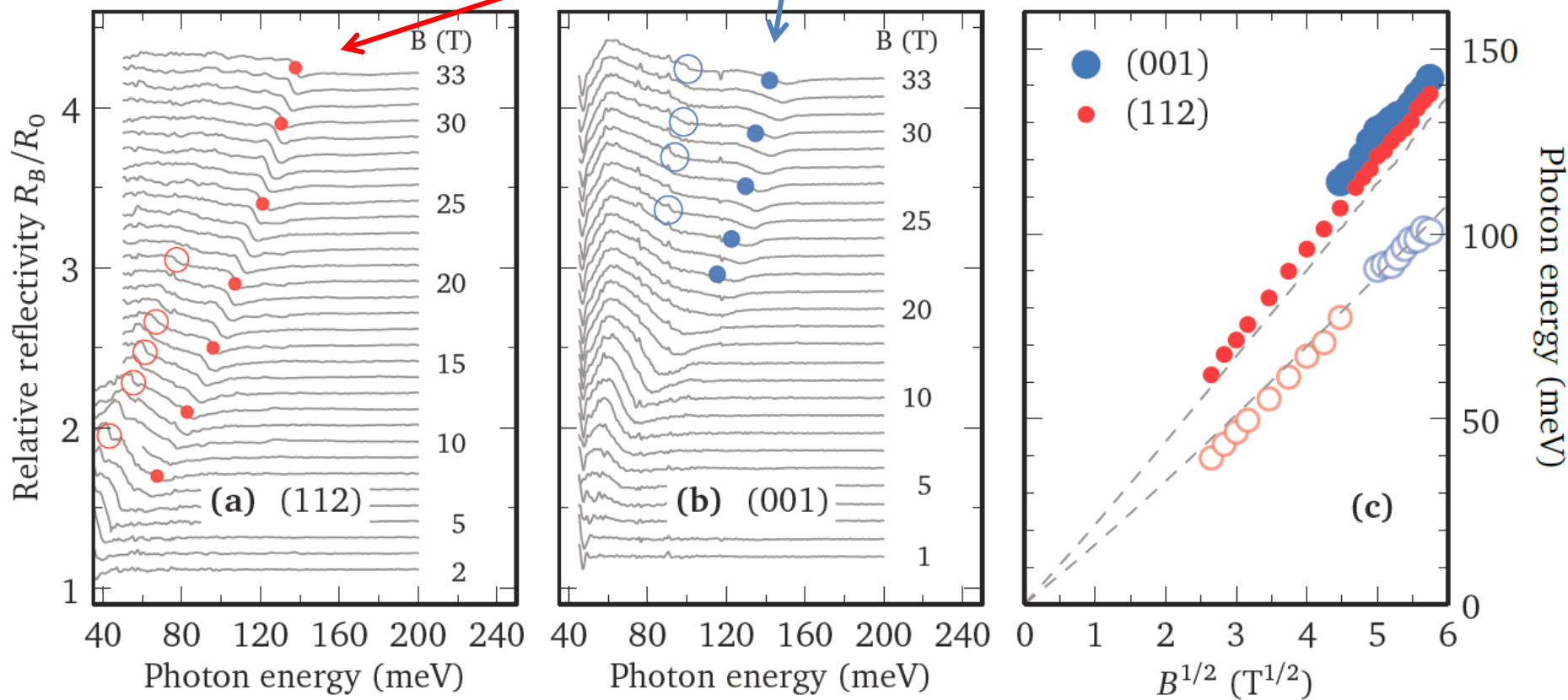


$$E_n = \pm v \sqrt{2e\hbar Bn + \hbar^2 k^2}$$

$$E_n = \pm v \sqrt{2e\hbar Bn}$$

Cd₃As₂ – High-field magneto-reflectivity

Cyclotron resonance (CR) in the quantum limit

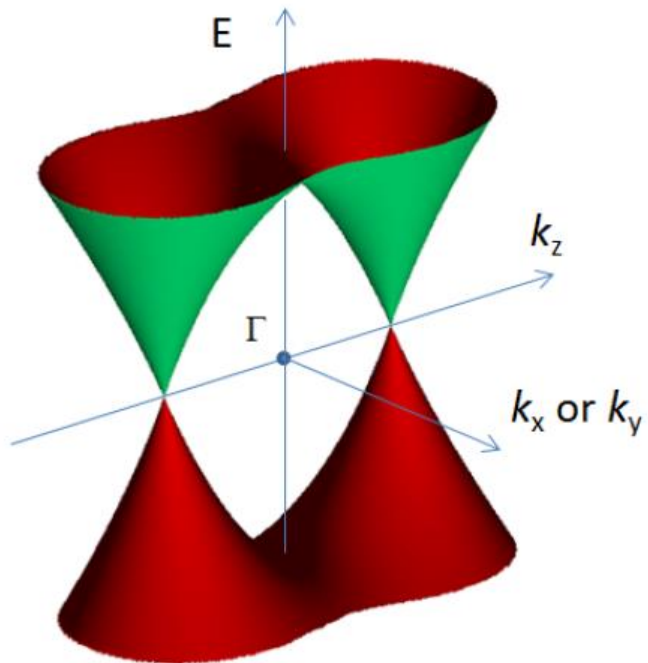


Magneto-optical response linear in \sqrt{B} = typical signature of massless particles

...massless yes, but not 3D Dirac

Cd_3As_2 – Electronic band structure

3D Dirac semimetal Cd_3As_2



How the band structure looks like?

What kind of massless carriers do we see?

Is Cd_3As_2 indeed a 3D Dirac semimetal?

Cd_3As_2 – Crystallographic structure

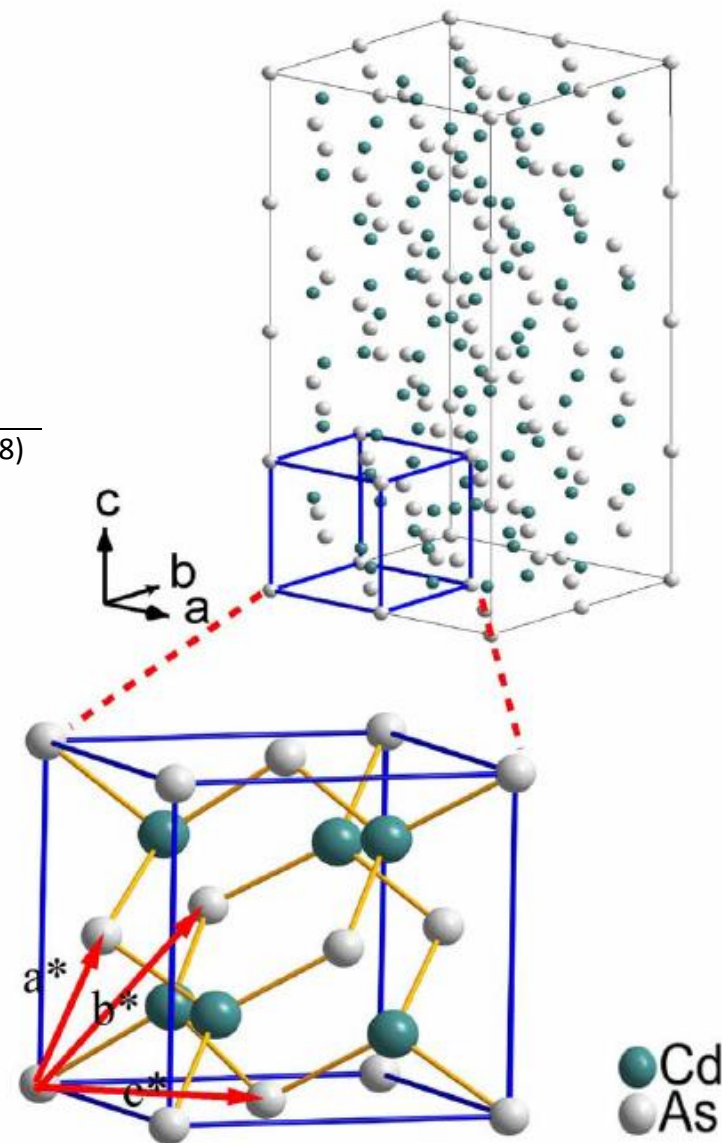
Tetragonal lattice with a large unit cell (~160 atoms)

G. Steigmann and J. Goodyear, *Acta Crystallogr. B* 24, 1062 (1968)

M. N. Ali et al. *Inorganic Chemistry* 53, 4062 (2014)

Ab initio calculations difficult for such a huge cell...

Z. Wang et al. *Phys. Rev. B* 88, 125427 (2013)



Cd_3As_2 – Crystallographic structure

Tetragonal lattice with a large unit cell (~160 atoms)

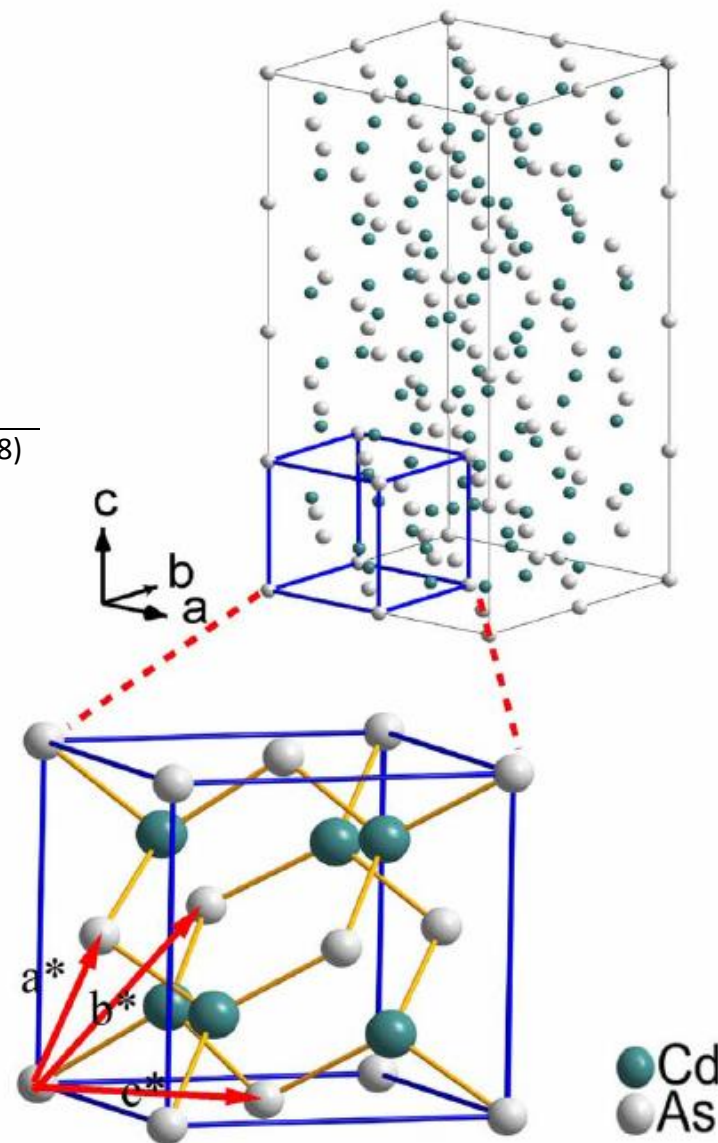
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Ab initio calculations difficult for such a huge cell...

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Composed from distorted cubic sub-cells
= “almost” zinc-blende lattice



Bodnar model of electronic bands in Cd_3As_2

BAND STRUCTURE OF Cd_3As_2 FROM SHUBNIKOV-de HAAS
AND de HAAS - van ALPHEN EFFECTS

J. BODNAR

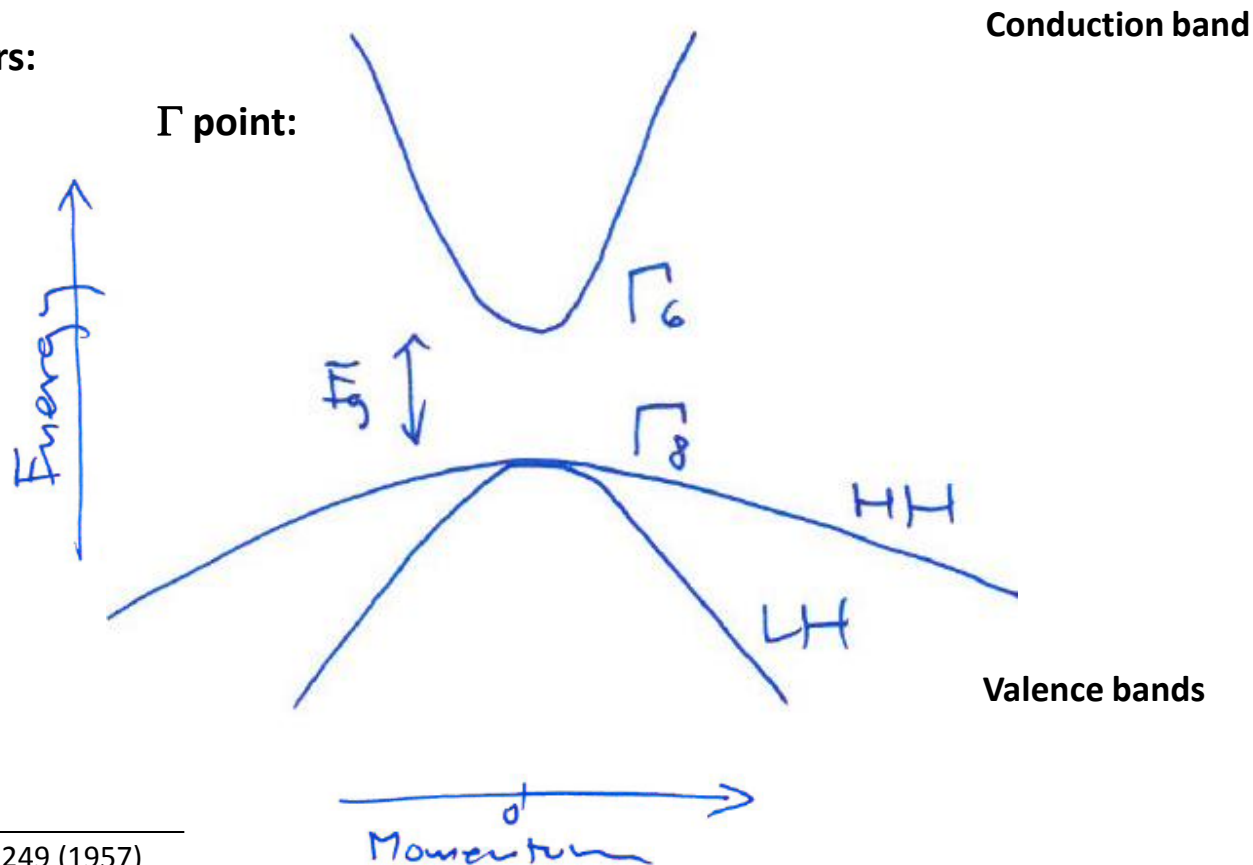
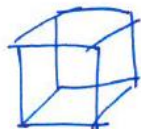
Department of Solid State Physics, Polish Academy of Sciences,
Zabrze, Poland

Experimental values of SdH and dHvA periods and cyclotron effective masses found by Rosenman and Doi et al. have been compared with the theoretical predictions derived in this work for a tetragonal narrow gap semiconductor. By the least square fit method the values of band parameters were obtained. It has been established that Cd_3As_2 has inverted band structure resembling HgTe under tensile stress.

Bodnar model of electronic bands in Cd_3As_2

Standard Kane model for zinc-blende semiconductors:

Cubic
(zinc-blende)

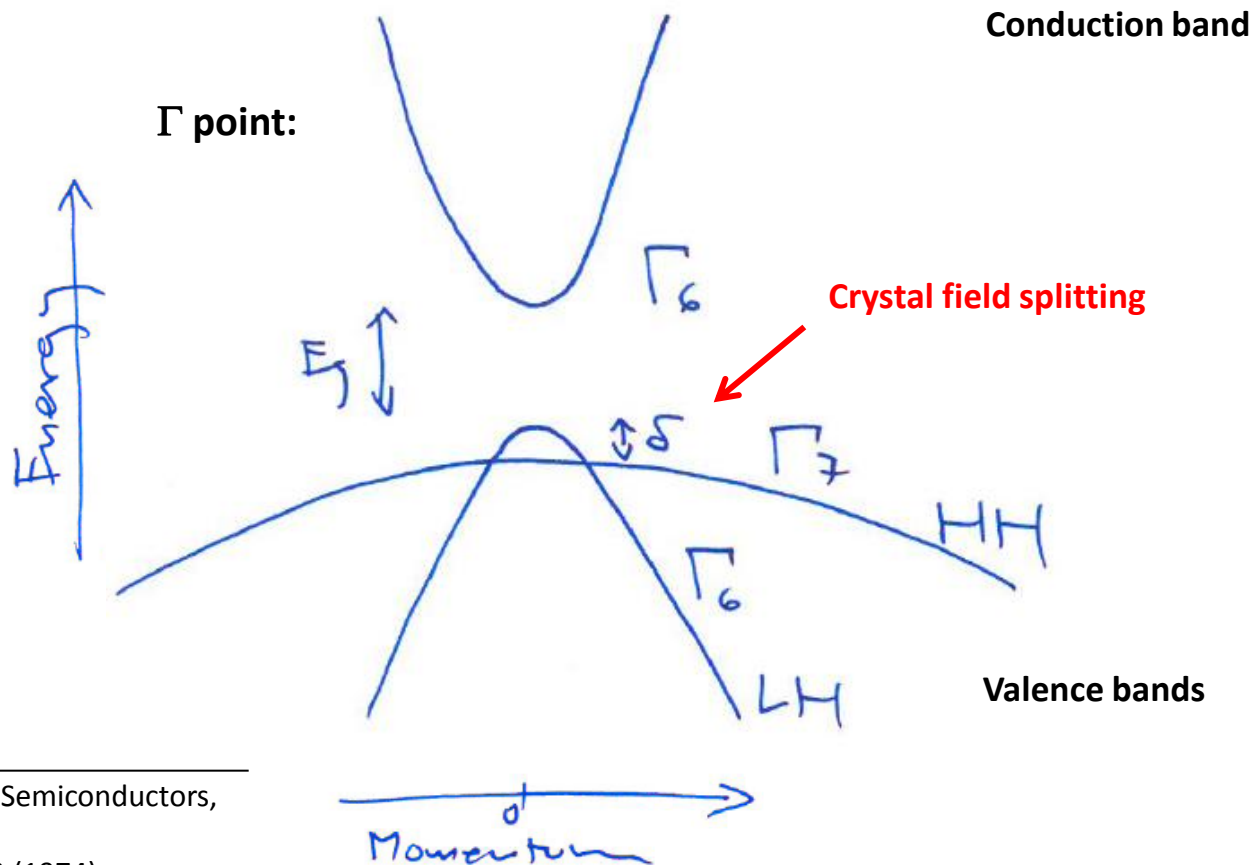
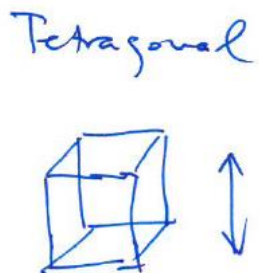


E. O. Kane, J. Phys. Chem. Solids 1, 249 (1957)

Valid for, e.g., GaAs, InAs, InSb, CdTe....

Bodnar model of electronic bands in Cd_3As_2

Kane model with a weak tetragonal distortion:

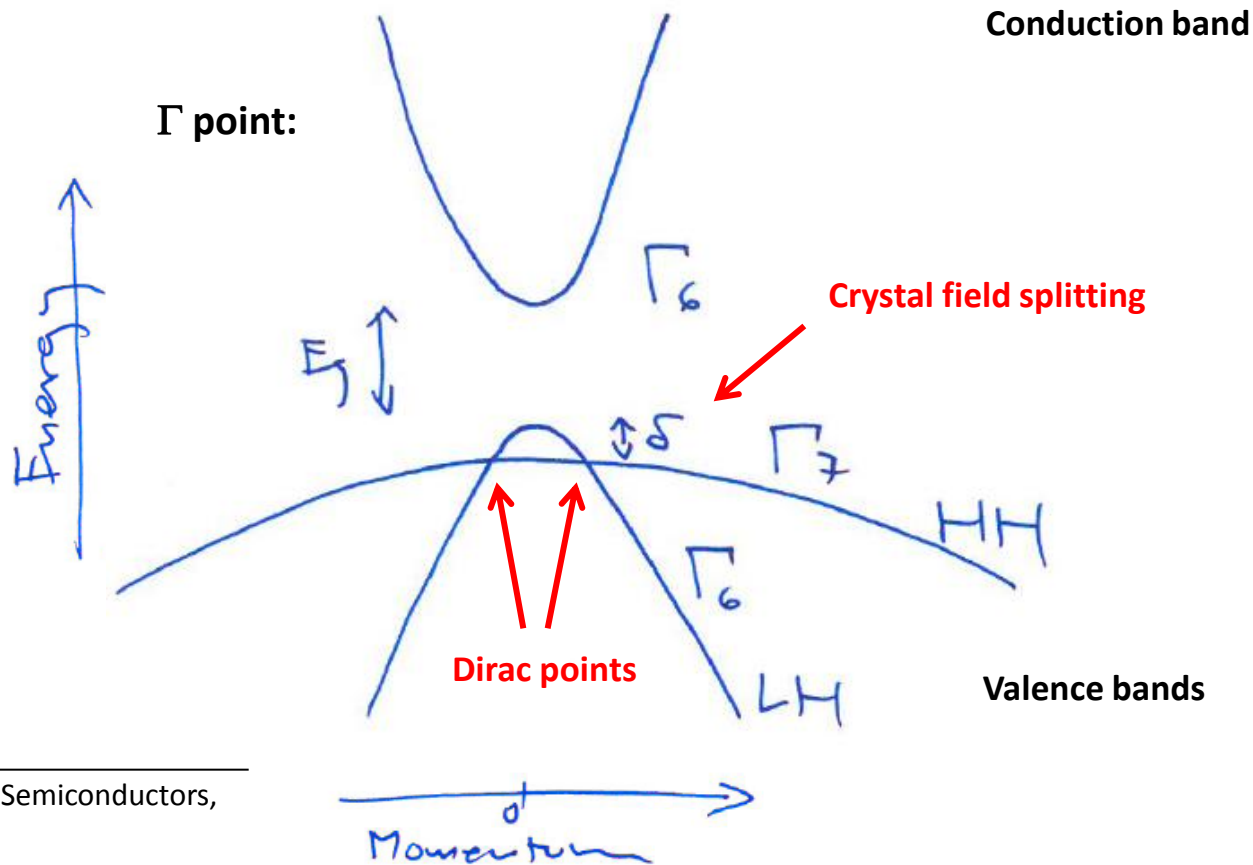
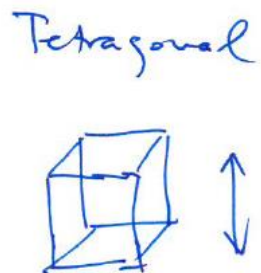


...along tetragonal axis

J. Bodnar, in Proc. III Conf. Narrow-Gap Semiconductors, Warsaw, (Elsevier, 1977) p. 311
 see also H. Kildal, Phys. Rev. B 10, 5082 (1974).

Bodnar model of electronic bands in Cd_3As_2

Kane model with a weak tetragonal distortion:

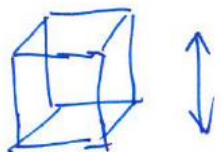


...along tetragonal axis

Bodnar model of electronic bands in Cd_3As_2

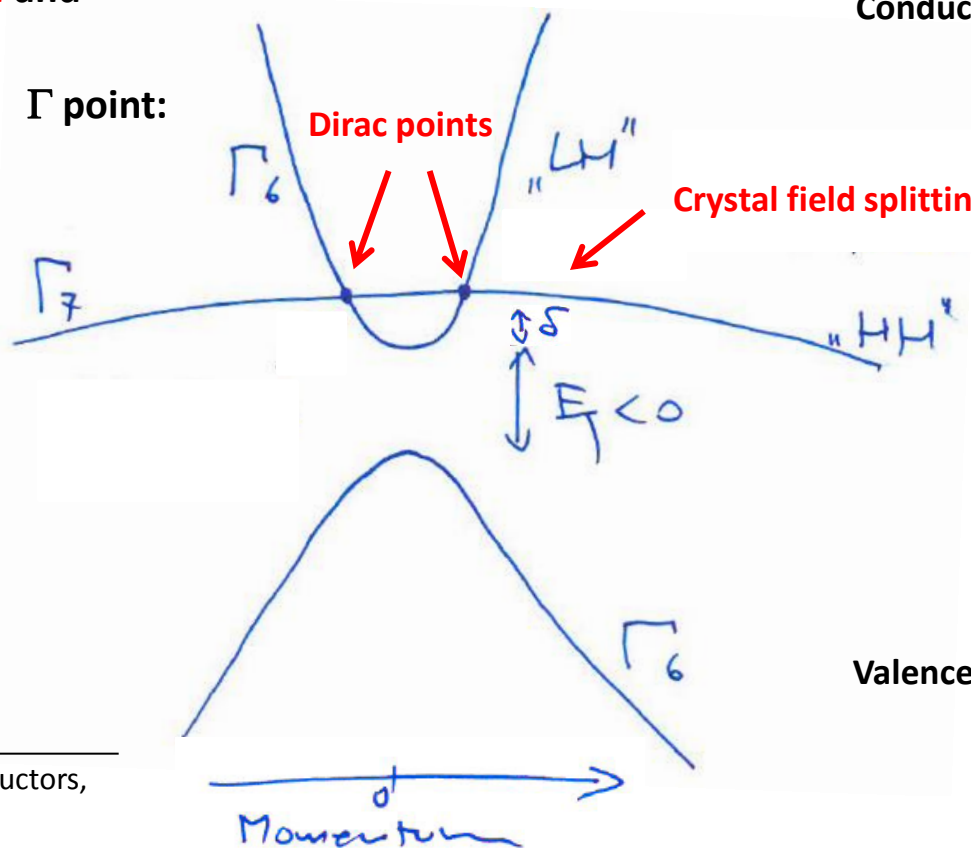
Kane model with a band **inversion** and a weak **tetragonal distortion**:

Tetragonal



Energy ↑

Γ point:



Conduction band

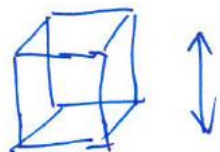
Valence bands

...along tetragonal axis

Bodnar model of electronic bands in Cd_3As_2

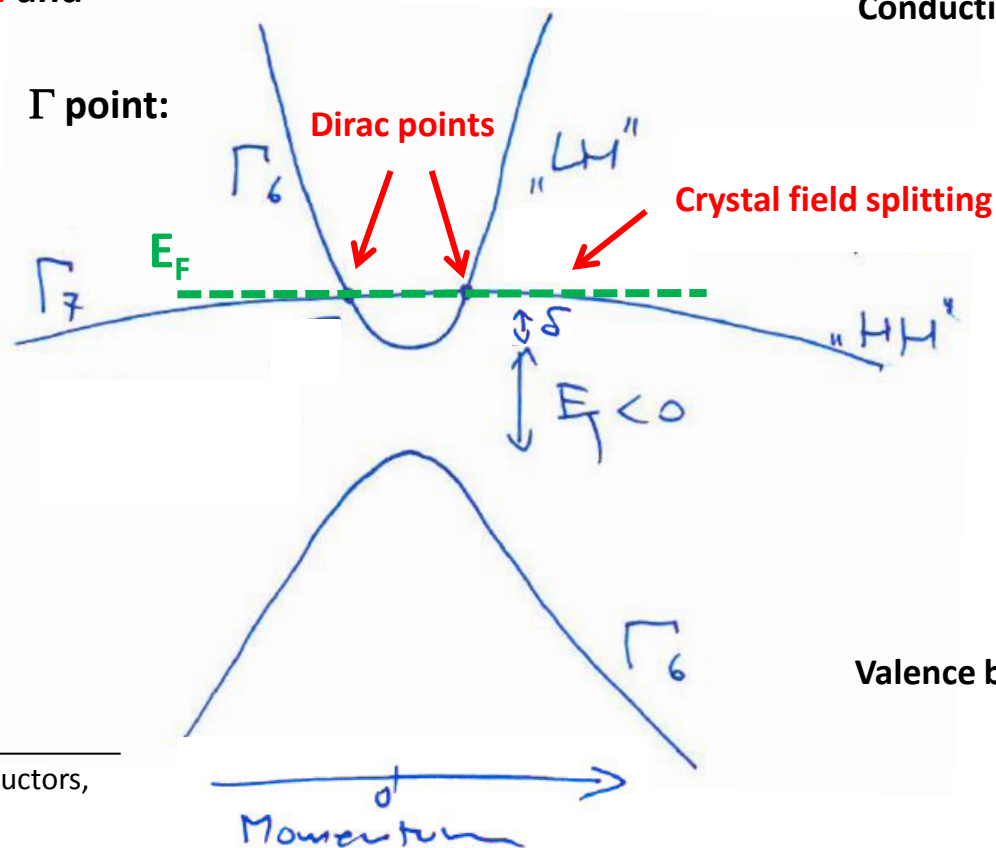
Kane model with a band **inversion** and a weak **tetragonal distortion**:

Tetragonal



Energy ↑

Γ point:



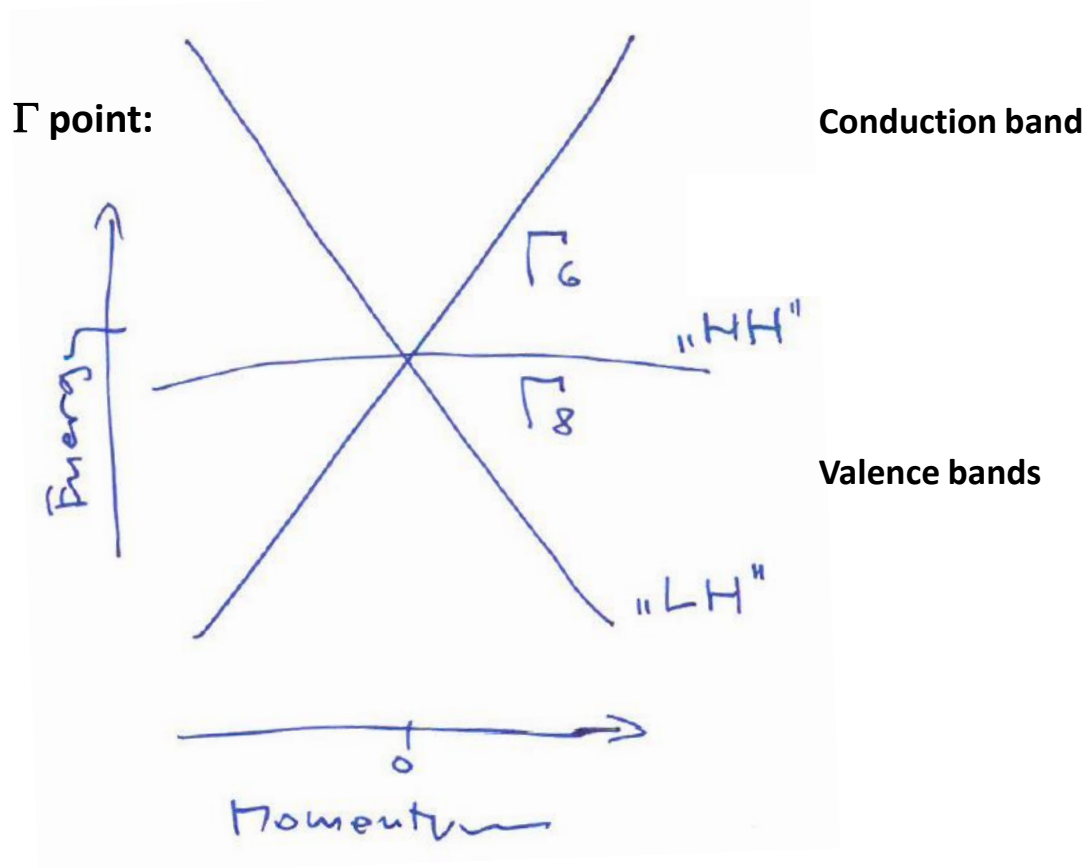
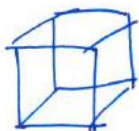
Conduction band

Valence bands

Bodnar model of electronic bands in Cd_3As_2

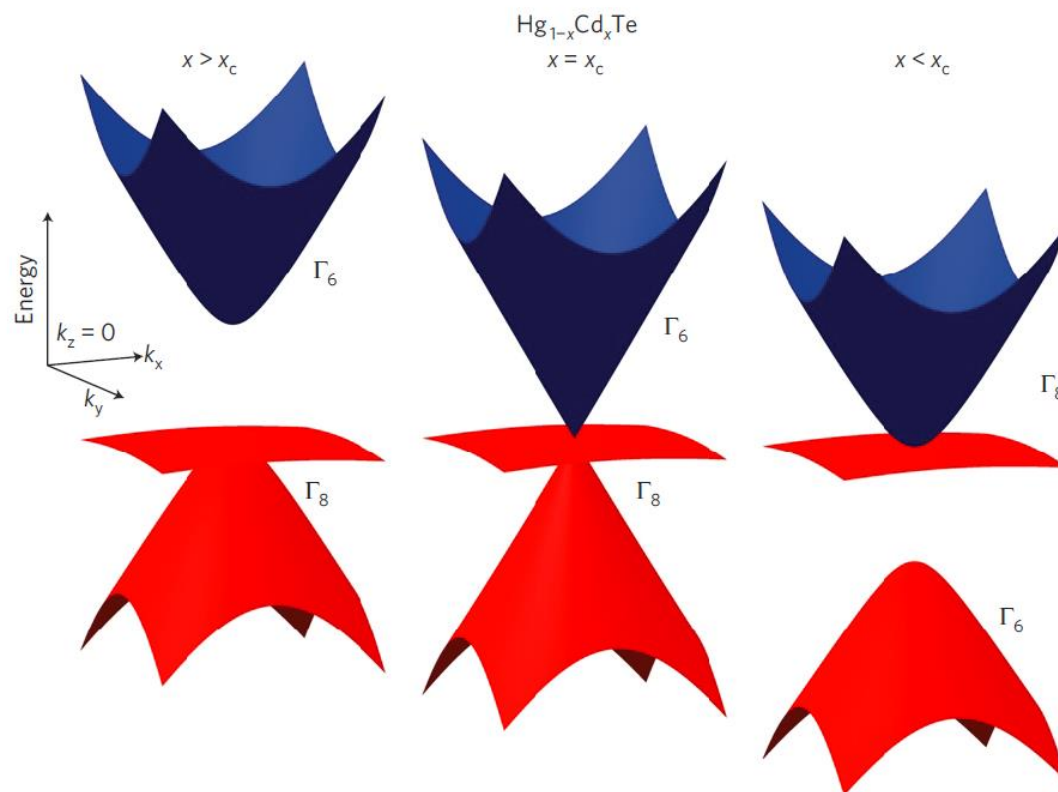
Standard Kane model
with a **vanishing band gap**:

Cube
(zone - bande)



E. O. Kane, J. Phys. Chem. Solids 1, 249 (1957)
 P. Kacman and W. Zawadzki, phys. stat. sol. (b) 47, 629 (1971)
 M. Orlita et al., Nature Phys. 10, 233 (2014)

Massless Kane electrons in gapless HgCdTe

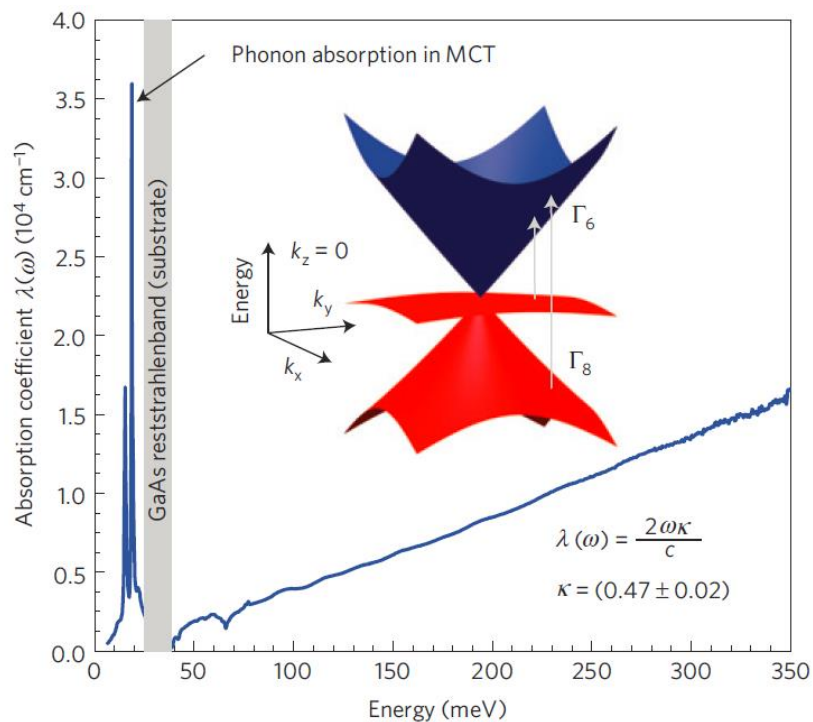


E. O. Kane, J. Phys. Chem. Solids 1, 249 (1957)
 P. Kacman and W. Zawadzki, phys. stat. sol. (b) 47, 629 (1971)
 M. Orlita et al., Nature Phys. 10, 233 (2014)

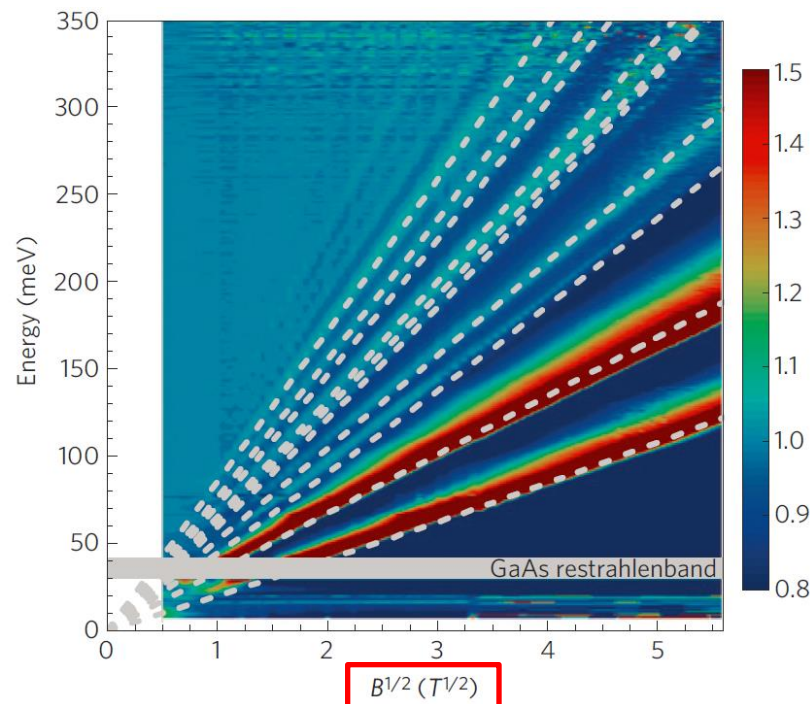
A single conical band in the center of Brillouin zone (due to accidental degeneracy of levels) hosting massless Kane electrons

Massless Kane electrons in gapless HgCdTe

Absorption coefficient linear in photon energy:



Magneto-optical response linear in \sqrt{B} :



Electronic bands in Cd_3As_2

Two energy scales of conical bands:

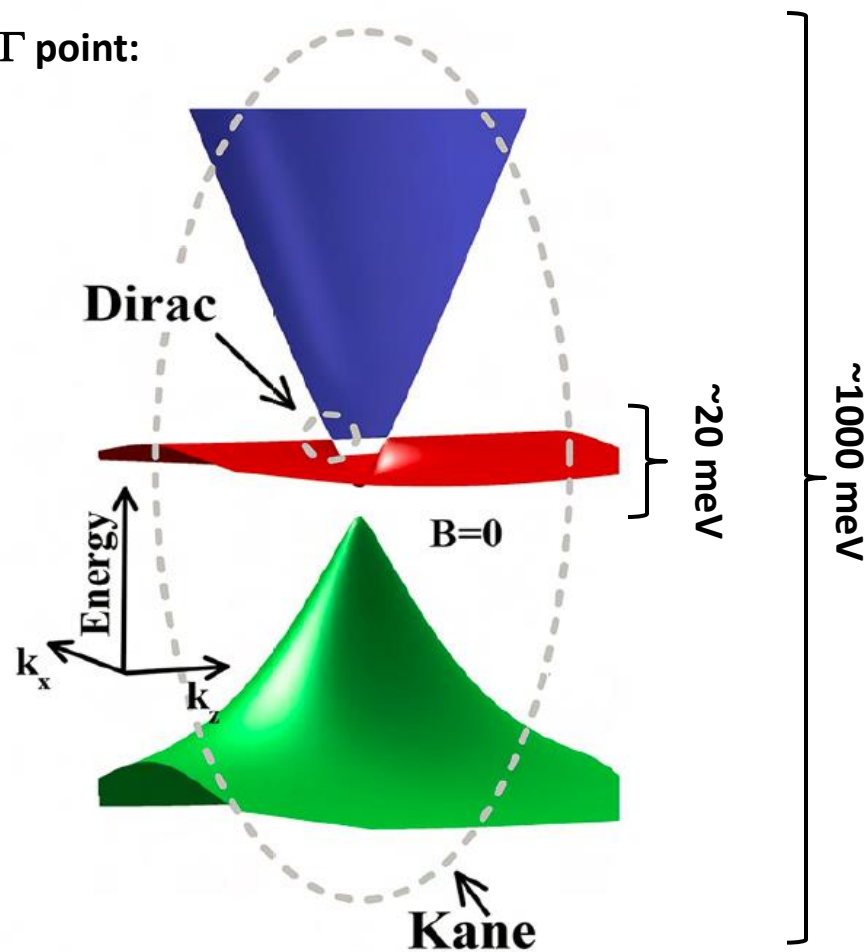
Two symmetry-protected **Dirac** cones at low energies

...due crossing of heavy and light hole band in a tetragonally distorted zinc-blende semiconductor

A single cone of massless **Kane** electrons, no symmetry protection

...appearing in zinc-blende semiconductors with a vanishing band gap

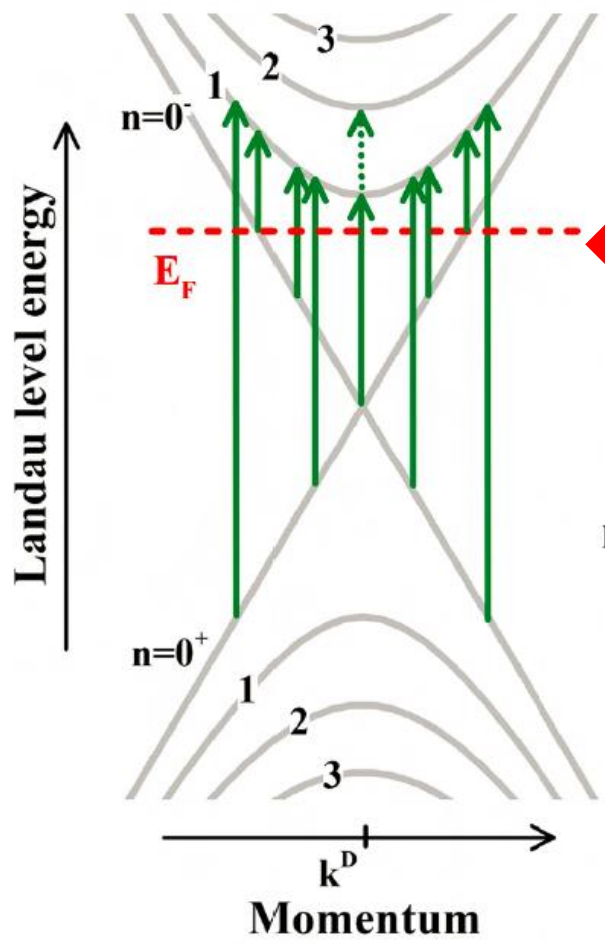
Γ point:



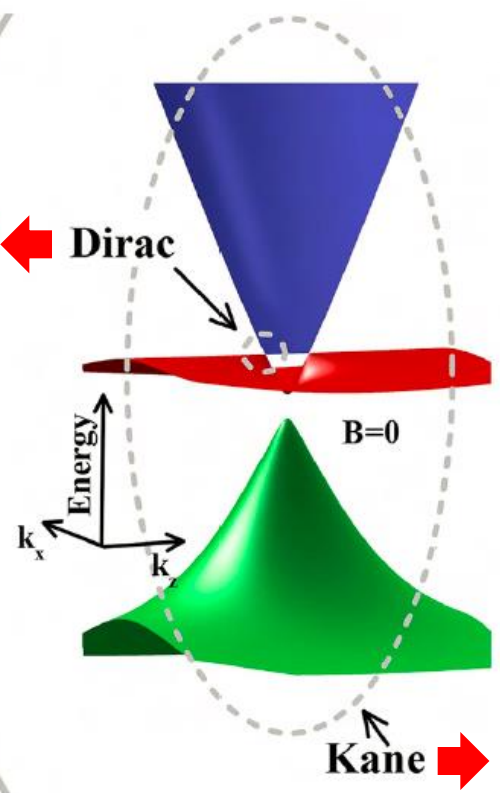
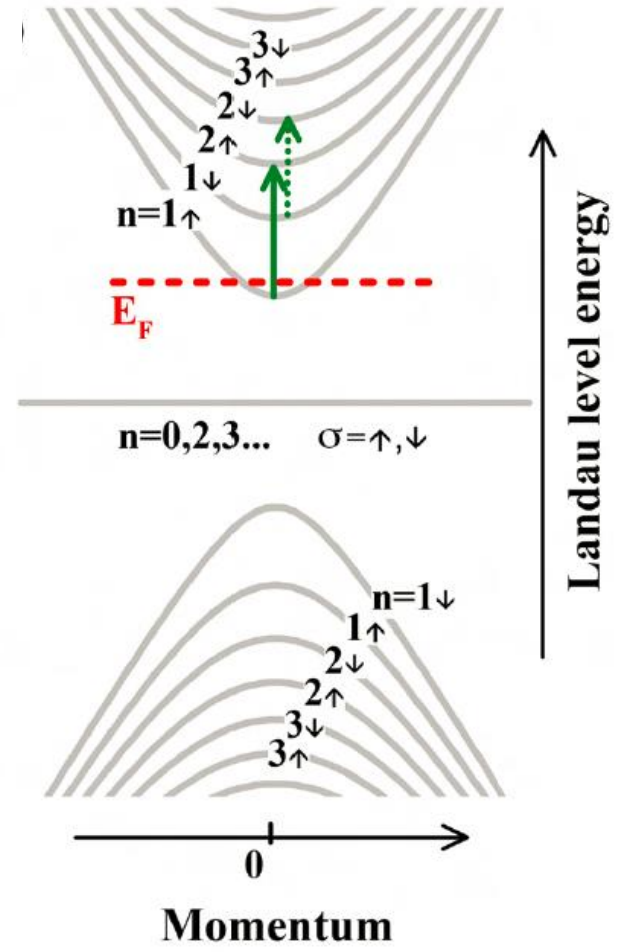
J. Bodnar, in Proc. III Conf. Narrow-Gap Semiconductors, Warsaw,(Elsevier, 1977) p. 311

3D massless Dirac/Kane electrons – Landau levels

Landau levels - Dirac electrons

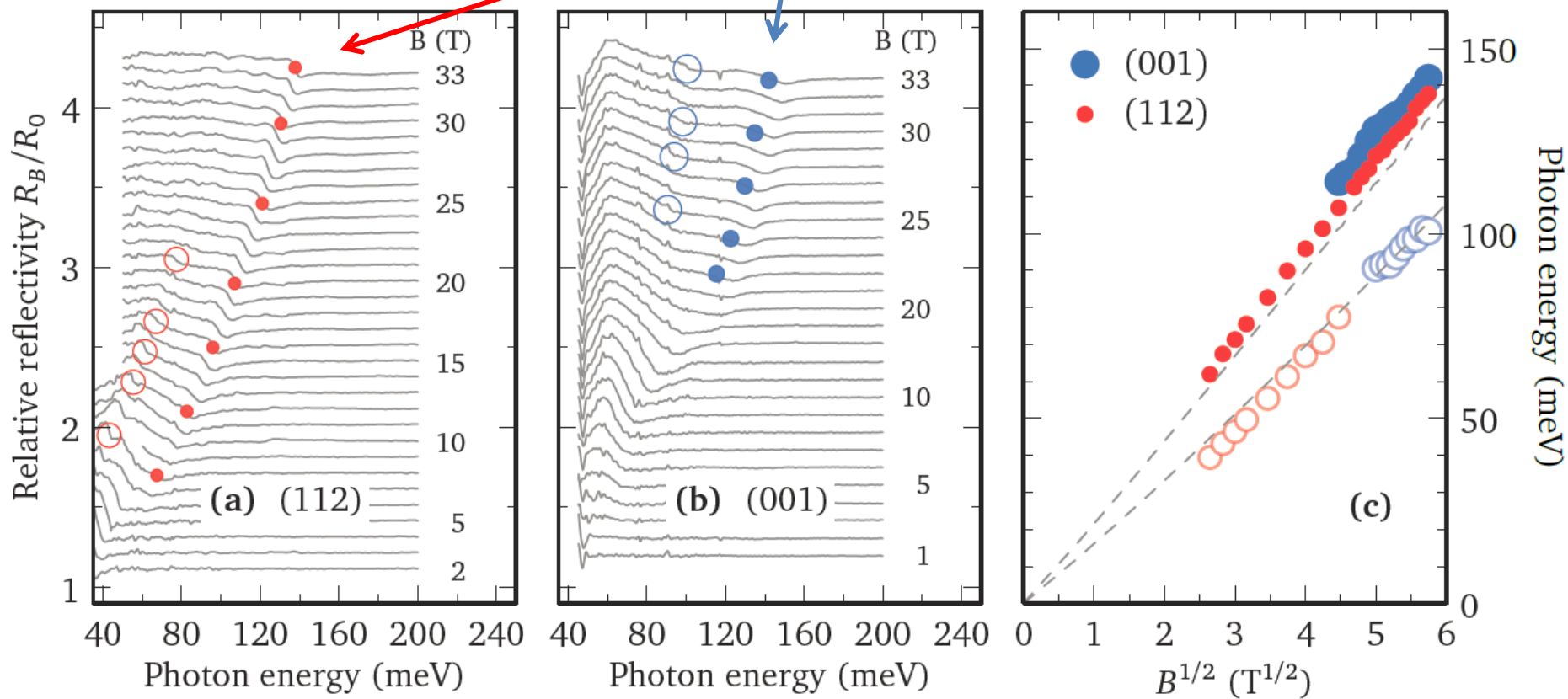


Landau levels - Kane electrons



Cd₃As₂ – High-field magneto-reflectivity

Cyclotron resonance (CR) in the quantum limit



Magneto-optical signature of **massless Kane electrons**

Conclusions

Discovery/fabrication of graphene triggered vast search for other materials with analogous properties

Silicene, germanene, topological insulators, molecular graphene, artificial graphene, semiconductors with giant Rashba-type spin splitting...

Materials with 3D conical bands reported

Na_3Bi , Cd_3As_2 , zero-gap HgCdTe , ZrTe_5 ,
 TaAs , NbAs

Z. K Liu et al., Science 343, 864 (2014)
 M. Orlita et al., Nature Physics 10, 233 (2014)
 Z. K. Liu et al., Nature Mater. 13, 677 (2014)
 S. Jeon et al., Nature Mater. 13, 851 (2014)
 R. Y. Chen et al., Phys. Rev. Lett. 115, 176404 (2015)
 B. Q. Lv et al., Nature Phys. 11, 724 (2015)
 L. X. Yang et al., Nature Phys. 11, 728 (2015)
 S.-Y. Xu et al., Nature Phys. 11, 748 (2015)

Cd_3As_2 is most likely a symmetry-protected 3D Dirac semimetal

...but only massless Kane electrons observed experimentally sofar

A. Akrap et al., arXiv:1604.00038 (2016)

Acknowledgements:

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