# Handedness resolved magneto-infrared spectroscopy of 2D and 3D Dirac materials



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20.01.17



Winter School New Frontiers in 2D materials: Approaches & Applications







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# Acknowledgments

#### University of Geneva

Iris Crassee Julien Levallois Jean-Marie Poumirol Pieter de Visser Ievgeniia Nedoliuk Michael Tran Dirk van der Marel Jérémie Teyssier

#### University of Zaragoza

Tetiana Slipchenko Luis Martin-Moreno

#### University of Michigan Ctirad Uher

#### LCNMI, Grenoble Milan Orlita

#### ETHZ (Zurich)

Peter Q. Liu Jérôme Faist

#### EPFL (Lausanne)

Michele Tamagnone Clara Moldovan Julien Perruiseau-Carrier Juan Mosig

#### nanoGUNE (San-Sebastian) Alexey Nikitin







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## Outline

Handedness resolved magneto-infrared spectroscopy

- Anomalous magnetic circular dichroism (MCD) and valley-polarized magneto-absorption in bismuth
- Electrically controlled terahertz MCD and Faraday rotation in graphene



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# Why handedness resolved IR/THz spectroscopy?

- Magnetic-circular dichroism (MCD) is a very important parameter to identify magneto-optical transitions
- Measuring MCD in the far-infrared/THz directly is very challenging (no broadband waveplates, or photoelastic modulators)



## Magneto-optical THz experiment in Geneva



- FTIR or time-domain THz spectroscopy
- Magnetic field up to 7 T (superconducting coil)
- Transmission/reflection
- Faraday rotation / Kerr rotation





## Magneto-optical Kramers-Kronig analysis (MOKKA)

#### Linear magneto-optical response

is determined by the transmission tensor:

or equivalently (in the circular basis):  $t_{\pm} = t_{xx} \pm it_{xy}$ 

$$\hat{t}(\omega,B) = \begin{pmatrix} t_{xx} & t_{xy} \\ -t_{xy} & t_{xx} \end{pmatrix}$$

#### We can directly measure:

Transmission (for linear polarized light)  $T = \frac{|t_+|^2 + |t_-|^2}{2}$ Faraday rotation:  $\theta_F = \frac{1}{2} Arg\left(\frac{t_-}{t_+}\right)$ 

Using a magneto-optical Kramers-Kronig analysis (MOKKA), we obtain:

Transmission (for circular polarized light):  $T_{+} = |t_{+}|^{2}$   $T_{-} = |t_{-}|^{2}$ Ellipticity/MCD

J. Levallois et al., Rev. Sci. Instrum. 86, 033906 (2015)

## **Basic assumptions for MOKKA**

- 1. Magneto-optical dielectric function  $\epsilon(\omega,B)$  is:
  - Iocal (q-independent)
  - $\clubsuit$  isotropic in the plane  $\perp$  B-field
- 2. Magnetic permeability  $\mu(\omega,B)$  is negligible
- 3. Magnetoelectric (multiferroic, axionic etc.) terms are negligible
- 4. B-field and light-propagation are both normal to the sample surface

## Parity relations for MO conductivity



Image from: http://blog.atlasrfidstore.com/choose-right-rfid-antenna

# Standard KK analysis of reflectivity



complex reflectivity (unknown) real reflectivity (known)  $r(\omega) = \sqrt{R(\omega)}e^{i\theta(\omega)}$  $R(\omega)$ known unknown Consider the function:  $f(\omega) = \ln r(\omega) = \ln \sqrt{R(\omega) + i\theta(\omega)}$ If f is analytic meromorphic e.g. or normal deflectivity in f bulk sample) then the KK relation applies:  $t_{O} = - m_{x} = t_{x} = t_{O} = - optics$ Since  $R(-\omega) = R(\omega) \implies \theta(\omega) = -\frac{\omega}{\pi} \wp \int_0^\infty \frac{\ln R(x)}{x^2 - \omega^2} dx + \pi$ This proves  $R(\omega)$  in (0, + $\infty$ ) determines <u>uniquely</u>  $r(\omega)$ the theorem:

For a reliable KK analysis one has to measure in a broad range and/or use additional measurements, such as ellipsometry

## MOKKA (direct integration)



Consider the function: unknown known  $f(\omega) = \ln\left(\frac{r_{-}(\omega)}{r_{+}(\omega)}\right) = \frac{1}{2}\ln c(\omega) + 2i\theta_{K}(\omega)$ 

where  $c(\omega) = \frac{R_{-}(\omega)}{R_{+}(\omega)}$ is circular-dichroism ratio

1<sup>st</sup> KK relation:  

$$c(\omega) = \exp\left\{\frac{8\omega}{\pi}\wp\int_{0}^{\infty}\frac{\theta_{K}(x)}{x^{2}-\omega^{2}}dx\right\} \implies R_{+}(\omega) = \frac{2R(\omega)}{1+c(\omega)} \implies R_{-}(\omega) = \frac{2c(\omega)R(\omega)}{1+c(\omega)}$$
2<sup>nd</sup> KK relation:  $\theta_{\pm}(\omega) = -\frac{1}{\pi}\wp\int_{-\infty}^{\infty}\frac{\ln\sqrt{R_{\pm}(x)}}{x-\omega}dx + \pi$ 

Since  

$$R_{\pm}(-\omega) = R_{\mp}(\omega) \qquad \Longrightarrow \qquad \theta_{\pm}(\omega) = -\frac{\omega}{\pi} \wp \int_{0}^{\infty} \frac{\ln \sqrt{R_{\pm}(x)R_{\pm}(x)}}{x^{2} - \omega^{2}} dx + \pi \mp \theta_{K}(\omega)$$

This proves the theorem:

 $R(\omega)$  and  $heta_K(\omega)$  in (0, + $\infty$ ) determine <u>uniquely</u>  $r_{\pm}(\omega)$ 

J. Levallois et al., Rev. Sci. Instrum. 86, 033906 (2015)

## Polarimetric experiment "fixed polarizer, rotating analyser" \*



Ellipsometry at normal incidence !

\* The same applies to the case «rotating polarizer, fixed analyzer»

## Extracting reflectivity and Kerr angle\*



## **Advantages of MOKKA**

- broadband (based on polarizers only)
- **4** can be applied at high magnetic fields (polarizers are fixed)

The first Faraday rotation spectroscopic measurement at 30 T



## **Advantages of MOKKA**



J. Levallois et al., Rev. Sci. Instrum. 86, 033906 (2015)

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## In the beginning was the Bismuth...





Image from: Z. Zhu et al. PRB 84, 115137 (2011).

Table I. Chronological table of phenomena discovered first in bismuth.

- Ultralow-density 3D-Dirac semimetal
- Strong spin-orbit coupling
- Huge band-mass anisotropy (> 200)
- Topological insulator when alloyed with Sb
- Year Discovery Diamagnetism (Brugmans; named by Faraday in 1845) 1778 Seebeck effect 1821 Nernst effect (Ettingshausen and Nernst) 1886 Kapitza's law of magnetoresistance 1928 1930 Shubnikov-de Haas effect de Haas-van Alphén effect 1930 1955 Cyclotron resonance in metals (Galt) 1963 Oscillatory magnetostriction (Green and Chandrasekhar)

#### Table from: Fuseya et al. JPSJ 84, 012001 (2015)

Numerous reviews are available...

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...

## **Bismuth crystals**

Very soft material, cleaved in liquid nitrogen, always on the trigonal plane



P.J. de Visser, J. Levallois et al. Phys. Rev. Lett. 117, 017402 (2016)

## Far-IR reflectivity + Kerr rotation on bismuth



Why not done before ?

- Spectroscopic (FTIR) experiments never combined with Kerr rotation
- Handedness-resolved experiments only at fixed (laser) wavelengths

# Handedness-resolved magneto-optical conductivity of bismuth (from MOKKA)



P.J. de Visser, J. Levallois et al. Phys. Rev. Lett. 117, 017402 (2016)

## Magnetic circular dichroism (MCD) in bismuth



Hugely suppressed for electrons !

P.J. de Visser, J. Levallois et al. Phys. Rev. Lett. 117, 017402 (2016)

## 3D Dirac gapped Hamiltonian and LLs in Bi

$$H_{e}(B) = \begin{bmatrix} \Delta & 0 & i\hbar v_{z}k_{z} & i\frac{\sqrt{2}}{l_{B}}\hbar va^{-} \\ 0 & \Delta & i\frac{\sqrt{2}}{l_{B}}\hbar va^{+} & -i\hbar v_{z}k_{z} \\ -i\hbar v_{z}k_{z} & -i\frac{\sqrt{2}}{l_{B}}\hbar va^{-} & -\Delta & 0 \\ -i\frac{\sqrt{2}}{l_{B}}\hbar va^{+} & i\hbar v_{z}k_{z} & 0 & -\Delta \end{bmatrix} v = (\Delta/m_{c})^{1/2}$$

$$v_{z} = (\Delta/m_{z})^{1/2}$$

$$l_{B} = (\hbar/eB)^{1/2}$$

$$u_{B} = (\hbar/eB)^{1/2}$$

$$u_{B}$$

Model agrees perfectly with Nerst, dHvA, SdH... effects Therefore LLs,  $E_F(B)$ , DoS(B) are for sure correct

> M. H. Cohen and E. I. Blount, Phil. Mag. **5**, 115 (1960). Z. Zhu et al. PRB **84**, 115137 (2011).

## 3D Dirac gapped Hamiltonian and LLs in Bi



**Experiment**: MCD = 13%

#### Does not explain the suppression of MCD !!!

## Mechanism of electron-MCD reduction in Bi



## Anisotropic mass as anisotropic metric

 $lpha~=~(m_x/m_y)^{1/2}$  - anisotropy parameter (~ 15 in Bi)

Mass anisotropy is equivalent  $\tilde{x} = \alpha^{-1/2}x$ ,  $\tilde{y} = \alpha^{1/2}y$ to anisotropic spatial metric  $\tilde{k}_x = \alpha^{1/2}k_x$ ,  $\tilde{k}_y = \alpha^{-1/2}k_y$ 

Anisotropic problem can be mapped isotropic problem:

$$\tilde{k}_x + i\tilde{k}_y \to \frac{l_B}{\sqrt{2}}\tilde{a}^+ , \, \tilde{k}_x + i\tilde{k}_y \to \frac{l_B}{\sqrt{2}}\tilde{a}^-$$

LL energies thus remain the same:  $\tilde{E}_n(B, k_z) = E_n(B, k_z)$ 

But LL wavefunctions are scaled :  $\tilde{\Psi}_n(B, x, y, k_z) = \Psi_n\left(B, \alpha^{-1/2}x, \alpha^{1/2}y, k_z\right)$ 

Magneto-optical conductivity (in one valley):

$$\sigma_{e1} = \begin{pmatrix} \alpha^{-1} s_{xx} & s_{xy} \\ -s_{xy} & \alpha s_{xx} \end{pmatrix}$$

## Adding three anisotropic pockets together...

 $\alpha =$ 

*m*,

В

Χ

 $\alpha = 14$ 

 $\alpha = 1$ 

One pocket conductivity:

$$\hat{\sigma}^{e1} = \begin{pmatrix} \tilde{\sigma}_{xx} & \tilde{\sigma}_{xy} \\ -\tilde{\sigma}_{xy} & \tilde{\sigma}_{yy} \end{pmatrix} = \begin{pmatrix} \alpha^{-1}s_{xx} & s_{xy} \\ -s_{xy} & \alpha s_{xx} \end{pmatrix}$$

### Add three pockets:

$$\hat{\sigma}^{tot} = \hat{\sigma}^{e_1} + \hat{\sigma}^{e_2} + \hat{\sigma}^{e_3} = \hat{\sigma}^{e_1} + \hat{R}\hat{\sigma}^{e_1}\hat{R}^{-1} + \hat{R}^{-1}\hat{\sigma}^{e_1}\hat{R}$$

$$MCD = \frac{\sigma_e^+ - \sigma_e^-}{\sigma_e^+ + \sigma_e^-} = \frac{2}{\alpha + \alpha^{-1}} \approx 13.8\%$$

Agrees perfectly with experiment !







- Elliptical light absorbed differently in different valleys
- Valley are selected by the azimuth angle



P.J. de Visser, J. Levallois et al. Phys. Rev. Lett. 117, 017402 (2016)

## Valley-selective absorption in TMDs

- Spin-polarized valence band (due to SO interaction)
- Valley-selective excitation with circularly polarised light



H. Zeng et al. Nature Nano. 7, 490 (2012)

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+ Handedness resolved magneto-infrared spectroscopy

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Electrically controlled terahertz magneto-optical phenomena in graphene



## Graphene field-effect transistor (g-FET) for THz wave manipulation



P. Q. Liu et al., Optica 2, 135 (2015)

## **Optical absorption in graphene**



Graphene membrane (visible)

bilayer

50

Absorption in graphene is 'universal': 2.3% but in the THz range it is much higher!!! (thanks to Drude carriers)

## Magneto-optical absorption of graphene: theory

B = 0





## Magneto-optical absorption of graphene: theory



\*interactions not included



- ~40% absorption (<1THz)</li>
- electron-hole symmetry



~40% absorption (<1THz)</li>

$$\sigma(\omega) = \frac{D}{\pi} \cdot \frac{i}{\omega + i/\tau}$$

- electron-hole symmetry
- Drude model works well



- electron-hole symmetry
- Drude model works well



- electron-hole symmetry
- Drude model works well



- electron-hole symmetry
- Drude model works well
- Scattering by charged impurities (decreases with n)

## B-field dependence of THz effects (high doping)



- Absorption >40% at 3 THz
- Cyclotron frequency is B-linear

## B-field dependence of THz effects (high doping)



- Absorption >40% at 3 THz
- Cyclotron frequency is B-linear
- Faraday rotation  $\theta_F$  >0.1 rad (6 deg)

Similar to epitaxial graphene on SiC: I. Crassee *et al.* Nature Phys.7, 48 (2011)

J.-M.Poumirol et al., Nature Communications, in press

## **Doping dependence of MO THz effects**



– Cyclotron frequency  $\omega_c \sim n^{-1/2}$ 

-  $\omega_c > 10$  THz can be reached !

Drastically different from 2DEGs!

## **Electric control of MCD and Faraday rotation**



- Magnetic circular dichroism and
   Faraday rotation can be inverted
   <u>electrostatically</u> !
- Electric control of a magnetic effect !
- Potentially new MO device functionalities !

Frequency (THz). Poumirol et al., Nature Comm. (2017) DOI: 10.1038/ncomms14626

## Graphene antidots: fabrication and transport



Square array of micron-size holes (antidots)

P. Q. Liu et al., Optica 2, 135 (2015)

## Graphene antidots: zero-field THz absorption



P. Q. Liu et al., Optica 2, 135 (2015)





J.-M.Poumirol et al., Nature Communications, in press



\*S.A.Mikhailov and V.A. Volkov, Phys. Rev. B 52, 17260 (1995) \*S.A.Mikhailov, Phys. Rev. B 54, 14293 (1996)

J.-M.Poumirol et al., Nature Communications, in press







#### Simulated field profiles in graphene antidots

### Faraday rotation in continuous and patterned graphene



Patterning blue-shifts frequency of maximum Faraday rotation

## Passive graphene-based THz elements

THz technology is lacking efficient passive devices:

- **4** Reciprocal: modulators, polarization converters etc.
- Non-reciprocal: isolators, circulators etc.

Graphene:

- Very efficient for manipulating THz waves
- Ultrathin (magneto-THz effects occurs in an atomically thin layer)
- Electrostatically switchable -> new functionalities

## **Optical isolator (diode, valve etc.)**



- Critical element in many applications
   (communications, lasers, optical computing etc.)
- Uses non-reciprocal magneto-optical effects
   (Faraday rotation or magnetic circular dichroism)
- Often very bulk (a few cm thick)
- Difficult to invert

Microwave (< 150 GHz) (based on ferrites) THz MIR-visible (> 15 THz) (1-10 THz) (based on magnetic glasses)



Nothing !!!



## First prototype THz graphene-based isolator



- insertion loss <7 dB</li>
- operates in reflection

M. Tamagnone et al., Nature Commun. 7, 11216 (2016)

## Reducing the B-field is imperative for applications

7 T



## **Summary**

**Handedness resolved magneto-infrared spectroscopy** J. Levallois et al., Rev. Sci. Instrum. **86**, 033906 (2015)

**4** Suppressed MCD and valley-polarized absorption in Bi

P.J. de Visser, J. Levallois et al. Phys. Rev. Lett. 117, 017402 (2016)

**4** Electrically controlled terahertz MCD and FR in graphene J.-M.Poumirol et al., Nature Communications, in press

**4** Towards graphene-based magneto-optical THz devices M. Tamagnone et al., Nature Commun. 7, 11216 (2016)

PhD and Postdoc positions available !!!





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