Ultrafast Probes for Dirac Materials

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Why Ultrafast Spectroscopy?

Ultrafast (10-100 fs) spectroscopy can resolve non-equilibrium dynamics (quasiparticle, transport etc.) at the fundamental time and spatial scales of electronic and nuclear motion.
Ultrafast Coherent Order Manipulation

Manipulation of order parameters ◆ Photoinduced phase transitions ◆ New non-thermally accessible phases.


DyFeO$_3$

T = 95 K

Faraday rotation (deg.)

Time delay (ps)

Vicario, Nature Phot. 2013


Above-bandgap excitation (arbitrary units)

Vibrational excitation

Fausti, Science 2011
Graphene: The Slice that Started It All

- **Graphene**: a basis for 0D buckyballs, 1D carbon nanotubes, and 3D graphite
- Quasiparticles are described by relativistic Dirac equation – *Dirac Material*
- Massless Dirac quasiparticles exhibit novel transport properties (high mobility, excellent conductivity)

Understanding the non-equilibrium behavior of photoexcited graphene is important for science and applications in detectors, solar cells and displays.

Bae et al. *Nat. Nanotech*. 2010

Bonaccorso et al. *Nat. Photonics* 2010
Quasiparticles in Graphene

Linear dispersion near Dirac point gives for relativistic quasiparticles:

\[ E \approx \hbar v_F k \quad E_{F}^{e,h} \sim \hbar v_F \sqrt{\pi N_{e,h}} \]

Are photoexcited quasiparticles in graphene relativistic too?

Two types of optical conductivity in graphene:

**Interband** is *constant* in a wide spectral range (flat 2.3% absorption)

**Intraband** differs for linear and parabolic bands

Measuring conductivity change after photoexcitation as function of \( N \) will indicate whether non-equilibrium quasiparticles are relativistic

We measure the photoinduced conductivity change:

$$\Delta \sigma = (\sigma_{\text{inter}} + \sigma_{\text{intra}})_{\text{Photo-excited}} - (\sigma_{\text{inter}} + \sigma_{\text{intra}})_{\text{Intrinsic doping}}$$

The change in conductivity, as measured in a visible pump-probe experiment, is dominated by the intraband component!
- 1.55 eV pump, 1.77 eV probe experiments
- Fermi energy after photoexcitation = 700 meV (for $N \sim 3.1 \times 10^{13}/cm^2$)
- Decay dynamics are qualitatively identical for all photon energies (1.74-2.42 eV)
- Electron-electron thermalization within <100 fs – Amplitude gives optical $\Delta \sigma$
- Electron-phonon thermalization within 1.4 ps
Hot Dirac Fermions in Graphene

Our experiment reveals the relativistic nature of photoexcited Dirac quasiparticles in graphene

Reflectivity (or conductivity) change follows \( \sqrt{N} \) from \( E_{F}^{e,h} \sim \hbar v_{F} \sqrt{\pi N_{e,h}} \)

Time-Resolved ARPES

High Harmonic Generation – Extreme nonlinear frequency upconversion

**STATIC ARPES:**
- probes electronic structure in both E and k domains

**DYNAMIC ARPES:**
- probes transient electronic structure changes in both E and k domains
- Fills excited states to reveal their structure

Photoexcited Fermi-Dirac Distribution in Graphene

- Is the Fermi-Dirac distribution of photoexcited carriers in graphene more like a metal (same $\mu_e$ and $\mu_h$) or like a semiconductor (separate $\mu_e$ and $\mu_h$)?

- Do processes like Auger recombination influence the dynamics at early times?

- Time-resolved photoemission experiments show that, in our samples, the photoexcited carriers retain separate F-D distributions for a few hundred femtoseconds.
Recombination of Electronic States in Graphene

- Ultrafast pump/probe experiment on CVD grown graphene
  - 30 fs IR pump and sub-10 fs, 30-eV probe via HHG
  - measure tr-ARPES
- A short-lived distribution of carriers and holes is formed after optical excitation.
- Separate populations are:
  - semi-conductor like ($\mu^* \neq 0$) at early delays
  - metallic like ($T^* \neq 0$) at later times
Topological Insulators

Materials with exotic surface states
- Linear $E-k$ dispersion
- TRS protection against scattering
- Locked spin-$k$ relationship
- Majorana Fermions
- Spintronics, optoelectronics

- Real materials are not ideal – dopants/defects result in significant bulk interference
- THz spectroscopy provides the ability to separate the collective motion of charge carriers in bulk vs. surface states

* after A. Lanzara
Optical Pump Terahertz Probe

35 fs @ 800 nm
1 KHz, 2.5W

chopper

beamsplitters

ZnTe
teflon

spherical lens

sample in cryostat

polarizing beamsplitter

photodiodes

lock-in

\[ E_{\text{sample}}(t) \]

\[ \sigma_{\text{real}}(\omega) \quad \sigma_{\text{imag}}(\omega) \]

conductivity (\( \Omega \cdot \text{cm}^{-1} \))

Frequency (THz)

Fourier Transform

Extraxt Complex Conductivity

Complex Transmissivity & Fresnel Analysis

\[ T(\omega) = \frac{E(\omega)_{\text{sample}}}{E(\omega)_{\text{ref}}} \frac{1+n_1}{1+n_1+\sigma(\omega)dZ_\omega} \]
Terahertz Conductivity of Bi$_2$Se$_3$

- **Low freq. spectra:**
  - Drude component: $1/\tau \sim 1$ THz
  - Bulk phonon: $\omega_0 \sim 1.9$ THz

- **Electron density consistent with**
  - $n_{surf} \sim 1.5 \times 10^{13}$ cm$^{-2}$

- **Drude term is thickness independent**
  - Surface.

- **Phonon is not → Bulk effect.**
Time-Resolved THz Spectroscopy

Fix THz gate delay at maximum and scan pump-probe delay

(a) 

$\Delta E/E_{max}$

Bi$_2$Se$_3$

10 QL 10 K

Pump-probe delay (ps)

(d) 

$\Delta E/E_{max}$

Pump-Probe Delay (ps)

4 $\mu$J/cm$^2$

6 $\mu$J/cm$^2$

10 $\mu$J/cm$^2$

24 $\mu$J/cm$^2$

50 $\mu$J/cm$^2$

100 $\mu$J/cm$^2$

200 $\mu$J/cm$^2$
Photo-Induced Conductivity in Bi$_2$Se$_3$

Drude-Lorentz Model:

Well described by **single** carrier type

Carriers in 20 QL decay faster

**Green**: Drude (free electron).

**Purple**: Phonon.
Photo-Induced Drude Properties in 20 QL

Low Fluence: increase scat. rate -> increase T

High Fluence: increase plasma freq. -> decrease T
At high fluence, phonon shifts - similar to increase in temperature.

Highest lattice temperature ~ 200 K
- Plasma frequency doesn’t change as much as in 20 QL sample.
- Scattering rate does, so the sample becomes more transparent at higher fluence.
Physical Picture

Phonon-induced bulk-to-surface scattering is not effective below $T_D=180\, K$

Thin 10 QL films are similar to graphene:
- Surface electrons dominate, but $\Delta \omega_p$ is small
- $\Gamma_{\text{surf}}$ increases due to e-h scattering and temperature rise ($\sim 200\, K$) due to e-ph relaxation

Thick 20 QL films:
- Surface response dominates at low fluences
- High fluences result in large number of bulk carriers $\Rightarrow$ higher $\Delta \omega_p$ and $\Gamma_{\text{bulk}}$
- Bulk electrons decay in $\sim 5\, \text{ps}$
- Surface electrons decay in 20 ps preserving high scattering rates


Hot surface carriers can be accessed independently from the bulk ones using THz spectroscopy

Topological Crystalline Insulators

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Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA
(Received 5 October 2010; revised manuscript received 31 December 2010; published 8 March 2011)

TI → Time Reversal Symmetry

TCI → Crystalline Symmetry

Metallic states on High Symmetry surfaces!

(001) surface

Dirac Point

Energy (eV)

$E(k)$ vs. $k$
Topological Phase Transition in $\text{Pb}_{1-x}\text{Sn}_x\text{Se}$

$P$-induced TPT in $\text{Pb}_{1-x}\text{Sn}_x\text{Se}$

Xi et al. PRL 113, 096401 (2014)
Topological Phase Transition in $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$

PbTe

Trivial

Doping-driven Topological phase transition

SnTe

TCI

$\text{Pb}_{1-x}\text{Sn}_x\text{Te}$

Can we use UOS to find the evidence for TPT with temperature and doping?

$X_c = 0.4$ at 5K

Yan et al. PRL 112, 186801 (2014)
Preliminary Results and Future Directions

Doping-induced TPT at 5 K

- Strong electron-phonon coupling in TI state – common to all TI
- Investigate the effect of magnetic field using THz spectroscopy to probe conductivity of photoexcited carriers.
- Apply circularly polarized pump to break TRS and study the dynamics of the $k$-spin locking process.

Temperature-induced TPT at $x=0.4$

- Coherent phonon
- Intervalley scattering
- e-ph coupling

<table>
<thead>
<tr>
<th>Temperature</th>
<th>Pb$<em>{0.8}$Sn$</em>{0.2}$Te</th>
<th>Pb$<em>{0.7}$Sn$</em>{0.3}$Te</th>
<th>Pb$<em>{0.6}$Sn$</em>{0.4}$Te</th>
<th>Pb$<em>{0.5}$Sn$</em>{0.5}$Te</th>
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<tbody>
<tr>
<td>5 K</td>
<td>Normalized $\Delta R/R$</td>
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$X_c$ ?
Temperature Dependence of Decay Amplitudes

Pb$_{0.6}$Sn$_{0.4}$Te

- $A_1 (10^{-5})$: Decreases with increasing temperature.
- $A_2 (10^{-5})$: Increases with increasing temperature.
- $A_3 (10^{-5})$: Slightly decreases with increasing temperature.
- $\tau_3$ (ps): Peaks around $T_c$.

$T_c$?