Nonlinear optical response and high-harmonic generation in 3D Dirac semimetals

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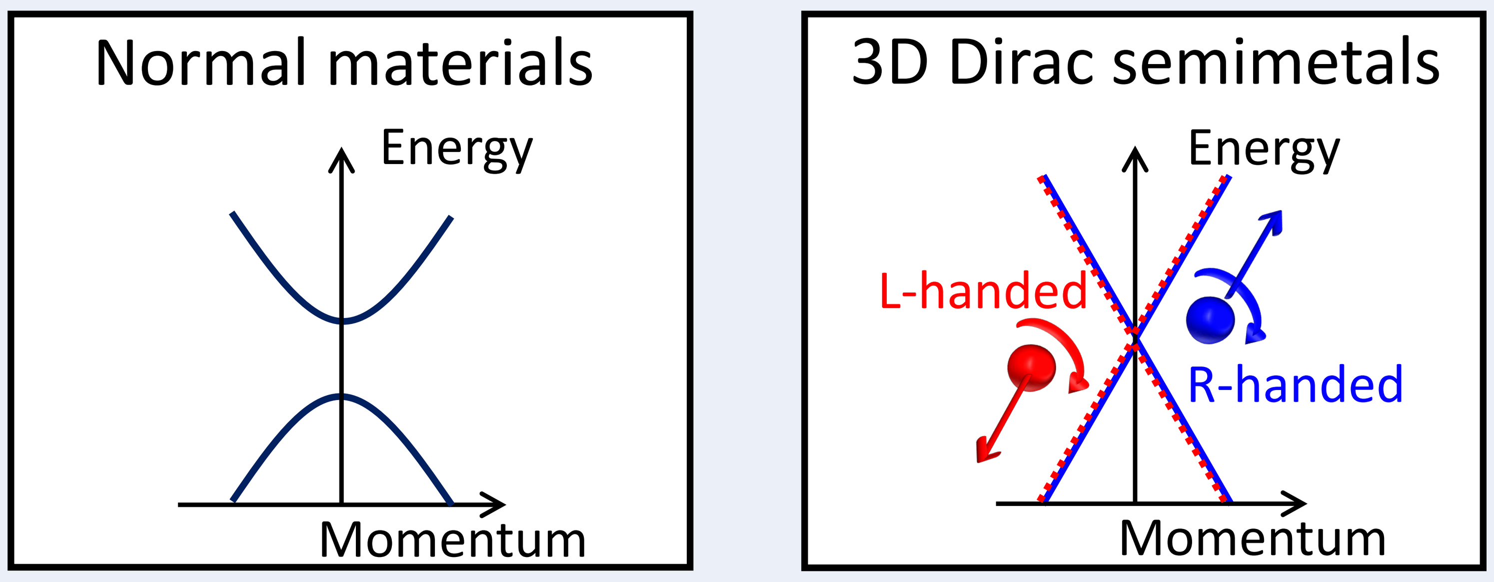
1 Introduction

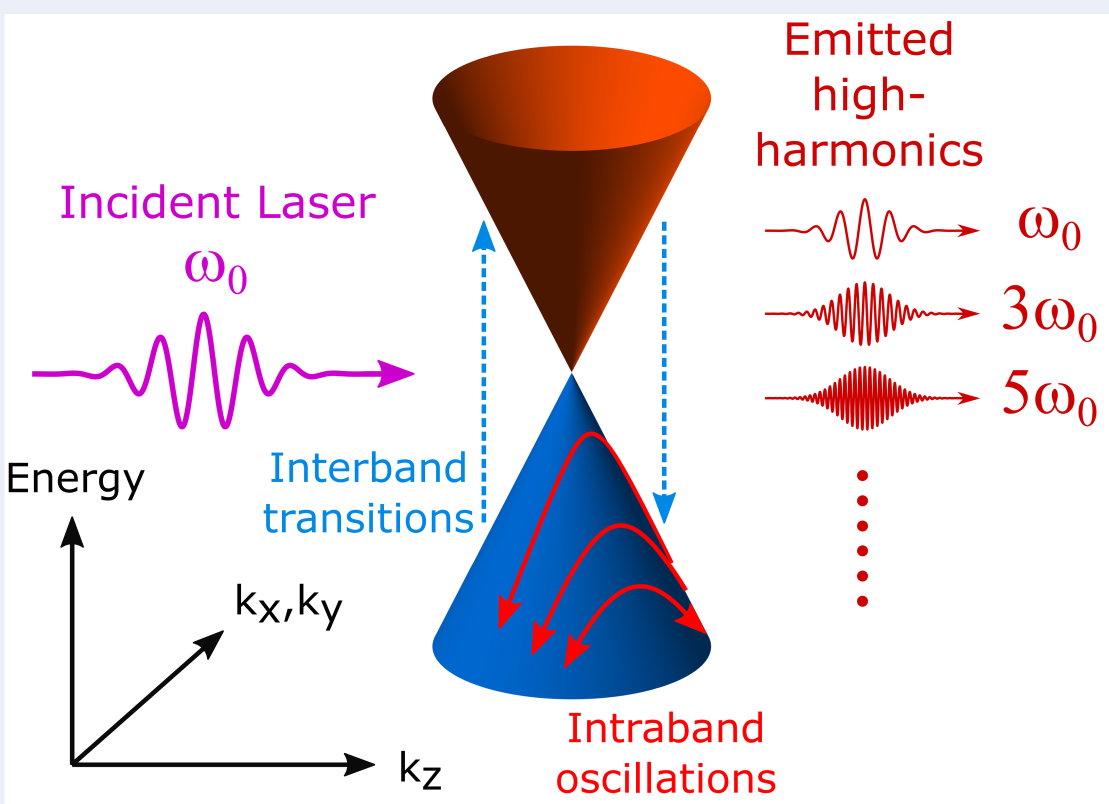
- 3D Dirac semimetals are 3D analogues of graphene.
- Its electrons obey a linear dispersion with a single gapless point and have chirality ("handedness").
- The linear dispersion leads to nonlinear field responses making them attractive for nanophotonic/plasmonic uses.



2 Motivation and objective

- The nonlinear field response of 3D Dirac semimetals imply efficient high harmonic generation (HHG).
- *Ab initio* simulations of real 3D Dirac semimetals are computationally expensive and obscure underlying physical mechanisms.
- Aim to obtain a simple and ***physically-transparent*** description of nonlinear field-response [1,2].
- Use our model to ***simulate electron dynamics*** in a ***computationally efficient*** way to ***predict HHG spectra***.

3 Theory and methodology



Schematic: laser-induced coupled interband & intraband currents generate odd-order high harmonics. The interband and intraband dynamics are coupled.

- Temporally propagate eigenspinors of chiral Weyl equations in momentum space to obtain a set of ODEs for band population evolution.
- ODEs cast into form of ***chiral Bloch equations*** which describe ***interband population dynamics*** and ***induced currents***.
- Fully ***non-perturbative***, ***anisotropic*** and no approximations beyond the ***single-electron massless regime***.

4 Key results

Chiral Bloch Equations:

$$\dot{\mathcal{N}}_R = -2\dot{\theta}\Re\left(\mathcal{P}_R e^{-2i\Omega}\right) + 2\dot{\phi}\sin\theta\,\Im\left(\mathcal{P}_R e^{-2i\Omega}\right)$$
$$\dot{\mathcal{P}}_R = i\mathcal{P}_R\dot{\phi}\cos\theta + \frac{1}{2}\mathcal{N}_R e^{+2i\Omega}\left(\dot{\theta} - i\dot{\phi}\sin\theta\right)$$

Right-handed electron

$$\dot{\mathcal{N}}_L = 2\dot{\theta}\Re\left(\mathcal{P}_L e^{-2i\Omega}\right) + 2\dot{\phi}\sin\theta\,\Im\left(\mathcal{P}_L e^{-2i\Omega}\right)$$
$$\dot{\mathcal{P}}_L = -i\mathcal{P}_L\dot{\phi}\cos\theta - \frac{1}{2}\mathcal{N}_L e^{+2i\Omega}\left(\dot{\theta} + i\dot{\phi}\sin\theta\right)$$

Left-handed electron

$$\phi = \arctan\left(\frac{v_y p_y}{v_x p_x}\right) \qquad \hbar\Omega = \int_{t_0}^{t} E(t')dt'$$
$$\theta = \arccos\left(\frac{v_z p_z}{\sqrt{v_x^2 p_x^2 + v_y^2 p_y^2 + v_z^2 p_z^2}}\right)$$

$\mathcal{N}$: Population difference between conduction and valence bands

$\mathcal{P}$: Interband coherence

Single-electron field-response:

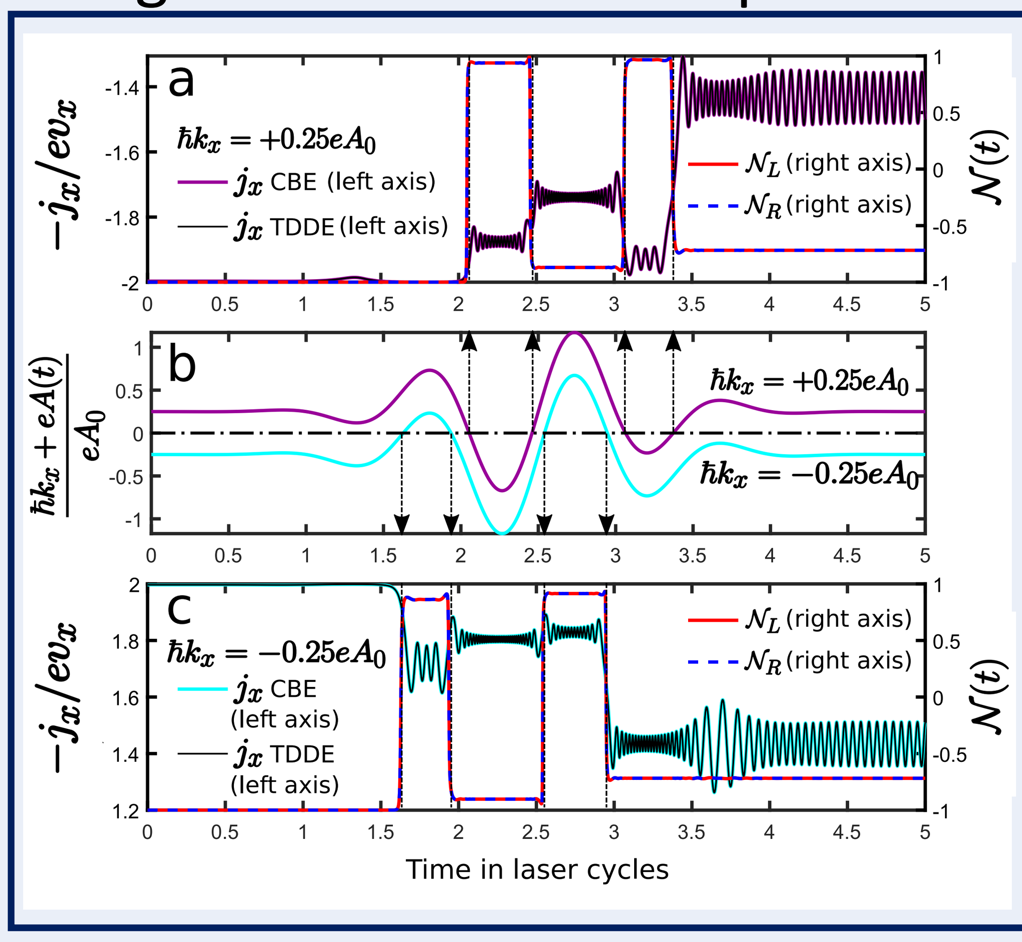


Figure 1: Time-evolution of (b) electron minimal-coupling momentum, and (a),(c) population difference (right axis) and single-electron current (left axis) for 2 starting momentum values. When the electrons are driven close to the Dirac point (horizontal dash-dotted line in (b)), instantaneous and complete population inversion occurs resulting in nonlinear current response.

Macroscopic field-response and Cd_3As_2 HHG spectra:

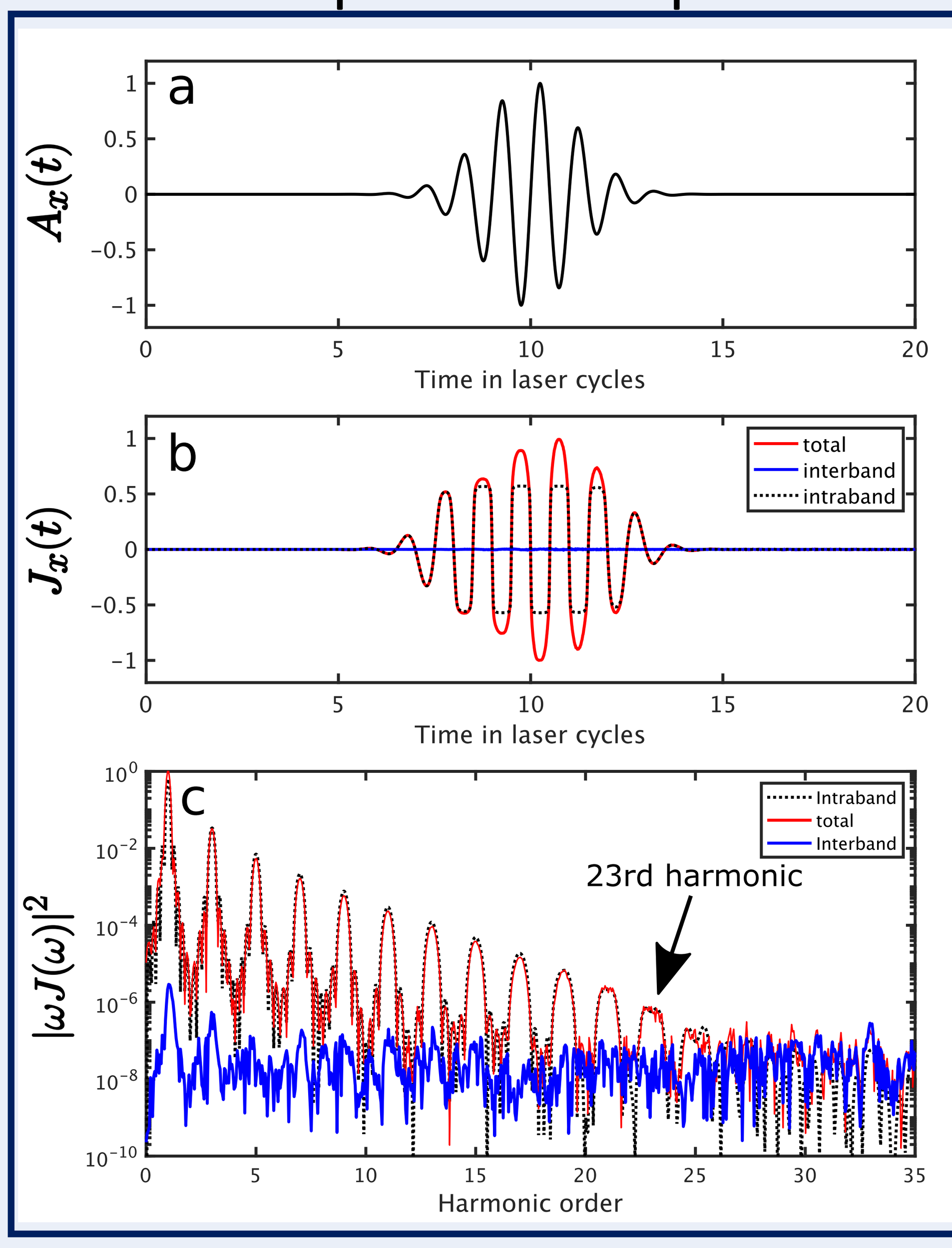


Figure 2: HHG in Cd_3As_2. (a) Vector potential temporal profile; (b) macroscopic current integrated over all momentum-resolved contributions; (c) HHG spectra obtained by performing a Fourier transform on the current. Odd-order high harmonics are dominated by the intraband current response. Interband response dominates for higher frequencies.

Cd_3As_2 parameters [3]:

$E_F = 250$ meV

$v_x = 1.28\times10^6$ m/s

$v_y = 1.3\times10^6$ m/s

$v_z = 3.27\times10^5$ m/s

$v_{x,y}$ higher than graphene's

Intraband linear conductivity derived using our formalism:

$$\sigma^{(1)} = \sigma_0 \frac{4E_f^2 g}{3\pi^2\hbar^2 v_f} \frac{\tau}{1 - i\omega\tau}$$

Result is in agreement with another work co-produced by our group [4].

References

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Acknowledgments

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