

# DOE Final Report

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Title: "THz Plasmonics and Topological Optics of Weyl Semimetals"

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## Accomplishments

THz magneto-optical properties of 3D topological Weyl semimetals were investigated with both THz spectroscopy and THz pump-probe measurements. The unique THz effects predicted in these materials may have important applications in THz technology. The electronic band structure was characterized spectroscopically through THz zero-field reflectance and/or cyclotron resonance measurements<sup>1,2</sup>. The studies included the dynamic chiral pumping and the study of the predicted novel magneto-electric effects arising from the underlying Berry curvature and magneto plasmonic-like effects in the absence of an applied magnetic field<sup>3,4</sup>. Chiral pumping in the extreme quantum limit was studied on Weyl semimetals to directly probe the chiral N=0 Landau level. Non-linear pump-probe measurements were used to measure the chiral pumping lifetime<sup>5</sup>.

Solids with topologically robust electronic states exhibit unusual electronic and optical transport properties that do not exist in other materials. A particularly interesting example is chiral charge pumping, the so-called chiral anomaly, in recently discovered topological Weyl and Dirac semimetals, where simultaneous application of parallel DC electric and magnetic fields creates an imbalance in the number of carriers of opposite topological charge (chirality). In an earlier study we investigated the Weyl metals Na<sub>3</sub>Bi and Cd<sub>3</sub>As<sub>2</sub><sup>1</sup>. In this grant we followed up with magneto-optical studies of TaAs, another Weyl semimetal<sup>2,4</sup>. More recently, we have also characterized other Weyl and Dirac systems that have come on line. The Physics community is still looking for the "hydrogen atom" of the Weyl semimetal. CoSi is one promising new system which features Weyl node spacing comparable to the Brillouin zone size. This system may be suitable for study of the predicted chiral plasmons that arise from the Berry curvature in Weyl materials. In other experiments gates can be applied to the samples in order to study the Fermi arc surface states by modulation reflectance spectroscopy at THz frequencies

## THz magneto-optical spectroscopy

Using THz optical spectroscopy and time resolved measurements on the Weyl semimetal TaAs in a magnetic field, we optically interrogated the chiral anomaly <sup>2,4</sup>.

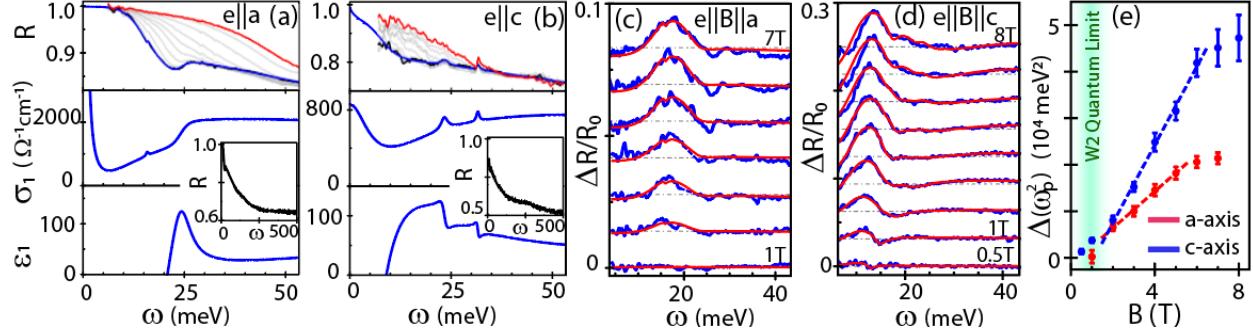


FIG. 1. (a) The reflectivity spectra  $R$  performed on a TaAs single crystal in the  $ab$  plane at zero field in the  $e \parallel a$  geometry. Multiple temperatures are reported that range from 300 K (red) to 10 K (black) with a superimposed fit (blue). The resulting low-temperature optical response functions are shown in the lower two panels. The inset shows the reflection taken over a broader spectral range. (b) Similarly, the zero-field reflectivity data and optical response functions are reported for the  $e \parallel c$  geometry in the  $ac$  plane. (c) Magnetic-field-induced changes in reflectivity  $\Delta R$ , normalized by the zero-field value  $R_0$ , are measured in the Voigt geometry on the as-grown  $ab$  facet with  $e \parallel B \parallel a$  in the  $ab$  plane at 10 K (blue) and fit (red). A dominant peak appearing near the screened plasma frequency is indicative of the chiral pumping effect. Spurious noise in a 2 meV band centered at 21 meV and replaced with a linear interpolation (thin blue dashed line). (d)  $\Delta R/R_0$  data are reported with  $e \parallel B \parallel c$  on a polished  $ac$  plane. (e) The enhanced Drude weight, reported as the change in the bare plasma frequency  $\Delta(\omega_p)$ , is derived from the fits in (c) and (d) and plotted as a function of magnetic field.

Chiral pumping was observed at THz frequencies in TaAs bulk single crystals for fields oriented along both the  $a$  and  $c$  axes<sup>2,4</sup>. Magnetoreflectance spectra of a TaAs bulk single crystal measured in Voigt geometries showed blueshifts of the screened plasma frequency with increasing magnetic field<sup>2,4</sup>. These blueshifts originate from Drude weight enhancements that are spectrally resolved from scattering rate effects. The enhancements agree with theoretical predictions of chiral pumping for TaAs. The Drude weight enhancement is accompanied by a reduction of the interband spectral weight in accordance with the  $f$ -sum rule, providing further confirmation of Drude enhancement<sup>4</sup>. The equivalence, of this transfer by the  $f$ -sum rule was theoretically used to derive the same chiral pumping effect predicted by other theoretical methods. The spectral observation of this transfer therefore strongly validates the interpretation that chiral pumping is responsible for the field dependence of the reflectance spectra. At higher fields, the Drude weight increases more slowly with field, which is interpreted as a decreasing Fermi velocity along the field. These results are shown to follow naturally from the  $f$ -sum rule, and offer an explanation for the observed increase in longitudinal magnetoresistance at high fields in transport experiments<sup>4</sup>.

Figure 2 shows the change in optical response in the measured spectral range obtained from the relative reflectance spectra and their Kramers-Kronig transformations for  $e \parallel B \parallel a$ . The frequency dependence of the large negative  $\Delta\sigma_1$  and the small negative  $\Delta\epsilon_1$  support the interpretation that the change in optical response is dominated by Drude weight enhancement<sup>4</sup>. The increasingly negative  $\epsilon_1$  at low frequency with field is indicative of a growing Drude weight. An increasingly negative  $\sigma_1$  at high frequency indicates loss of interband spectral weight. Therefore, these two graphs show a transfer of spectral weight from high to low frequency. The increase of the Drude spectral weight at very low frequency is not directly observable since the scattering rate is small compared to the lowest measured frequency. This interpretation is consistent with the fact that  $\Delta R(B)/R_0$  for  $e \parallel B \parallel a$  is fitted well by increasing the Drude weight of the zero-field optical response<sup>4</sup>.

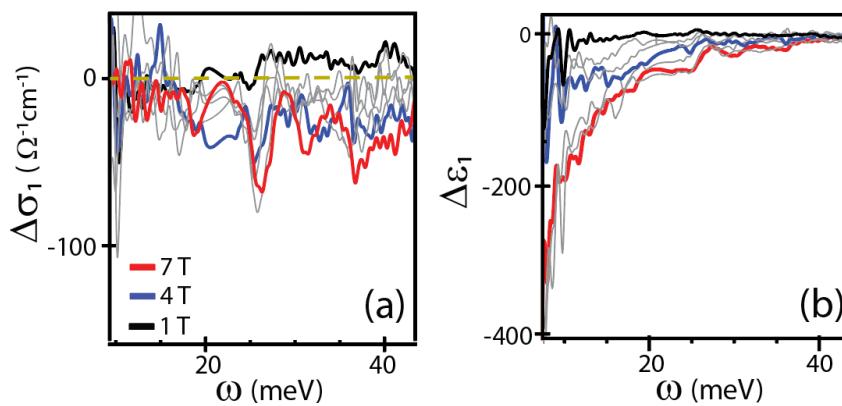


Fig. 2. Shown in (a) and (b), are  $\Delta\sigma_1$  and  $\Delta\epsilon_1$  for  $e \parallel B \parallel a$  obtained using  $\delta R/R_0$  and its KK-transformation and the zero-field dielectric function. Drude enhancement manifests mainly as  $\Delta\epsilon_1$ .  $\Delta\sigma_1$  is dominated by changes to the optical response of the Drude and IBTs. A dark yellow dashed line is included in (b) to show that  $\Delta\sigma_1 < 0$  in the measured spectral range, consistent with loss of IBT spectral weight. The change in optical response is dominated by  $\Delta\epsilon_1$ .

### Pump Probe experiments

We studied the concept of the dynamical chiral charge pumping and its optical control in a Weyl semimetal<sup>5</sup>. See Fig. 3. From the pump-probe results at zero magnetic field we conclude that the response is attributed to the pump-induced hot carriers in TaAs. We showed the pump-probe measurement results at various magnetic fields applied parallel to the pump-probe polarization, and for different pump fluences<sup>5</sup>. Using a simple model for Weyl nodes and nonlinear magneto-optical response, we obtained the pump-induced change in optical conductivity. Our experimental and theoretical results illustrate the ability to optically control the dynamical chiral charge pumping. While there is no static current produced by the pump pulse, it creates correlated excitations (or quasiparticles) in the opposing Weyl nodes that persist long after the pump pulse has passed. See Fig. 4. These quasiparticles alter the reflectivity of a subsequent weaker probe pulse in a measurable way. The lifetime associated

with this nonlinearity is the dynamical chiral charge relaxation time and is measured to be  $1 \text{ ns}^5$ . See Fig. 4. We further confirmed our results by showing that, for the case in which the pump polarization is perpendicular to the applied magnetic field, no long-lived response associated with nonlinear dynamical chiral charge pumping is observed<sup>5</sup>. Our findings paved the way for future studies exploring optical control of topological transport phenomena in solids, and investigating long-lived Chiral Weyl fermions for quantum optoelectronic applications.

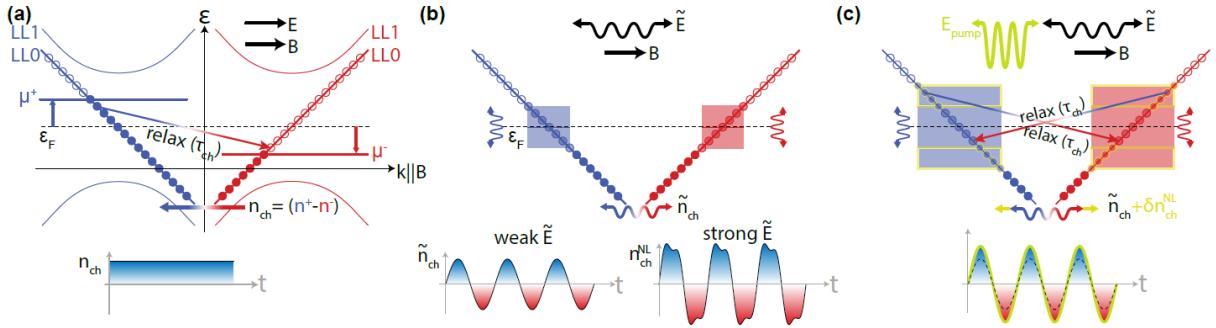


FIG. 3. (a) Static charge pumping of chiral carriers in the zeroth Landau level. Red and blue colors denote different chiralities of carriers. The chiral charge imbalance relaxes back to equilibrium with a time constant of  $\tau_{ch}$ . (b) Dynamical charge pumping, shaded blue and red regions around Fermi level  $\varepsilon_F$ , are produced by an oscillating optical field polarized parallel to  $B$ . For strong optical excitation, the chiral charge oscillation between the Weyl nodes becomes nonlinear in  $\tilde{E}$  leading to anharmonic oscillation of chiral charges. (c) Strong optical field  $\tilde{E}$  pump enhances the chiral current generated by  $\sim E \parallel B$ . When  $E_{pump}$  is turned off, the extra pump-induced chiral carrier distribution (illustrated by a yellow border) slowly relaxes back via inter-Weyl-node relaxation.

Applying parallel static electric  $E$  and magnetic  $B$  fields produces a chiral current  $J_{ch}$  that pumps the carriers from one Weyl node to the other with opposite chirality and unbalances the chiralities of the carriers. The chiral charge imbalance relaxes back to equilibrium with a time constant of  $\tau_{ch}$ . (b) Dynamic charge pumping of chiral carriers in the zeroth Landau level: oscillating optical field polarized parallel to a magnetic field  $B$  causes harmonic oscillation of number of chiral carriers at the same frequency. This generates a distribution of chiral carriers around the equilibrium Fermi level  $E_F$  shown by shaded blue and red regions. For strong optical excitation, the chiral charge oscillation between the Weyl nodes becomes nonlinear in  $\sim E$  leading to anharmonic oscillation of chiral charges. (c) Nonlinear optical control of dynamic chiral anomaly: strong optical field  $\sim E_{pump}$  enhances the chiral current generated by  $\sim E \parallel B$ . When  $\sim E_{pump}$  is turned off, the extra pump-induced chiral carrier distribution (illustrated by a yellow border) slowly relaxes back via inter-Weyl-node relaxation.

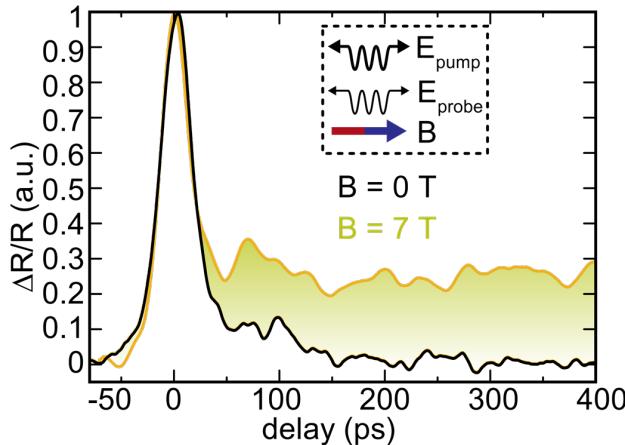


Fig. 4. (a) Pump-induced increase in probe reflection for  $B = 0$  T (black) and  $B = 7$  T (yellow) in arbitrary units as a function of pump-probe time delay. The peak around 60 ps is caused by a replica—the reflected pump pulse from the cryostat window.

This research has continued in magneto-optical studies of CoSi, another topological semimetal.

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4. *“Optical evidence of the chiral magnetic anomaly in Weyl semimetal TaAs”*  
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5. *“Nonlinear Optical Control of Chiral Charge Pumping in a Topological Weyl Semimetal”* M. Mehdi Jadidi, Mehdi Kargarian, Martin Mittendor, Yigit Aytac, Bing Shen, Jacob C. Konig-Otto, Stephan Winnerl, Ni Ni, Alexander L. Gaeta, Thomas E. Murphy, and H. Dennis Drew, *Physical Review B*, 102, 245123 (2020).