Supplementary Online Information for "Chiral plasmons without magnetic field"

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Current density and the conductivity tensor

For a complementary view to the approach employed in the main text, here we present an alternative formulation of the plasmon equations of motion. Rather than focusing on the semiclassical equations of motion for the particle and velocity density fields, here we work with the charge density $\rho(\mathbf{r},t) = -en(\mathbf{r},t)$, the current density $\mathbf{j}(\mathbf{r},t)$, and the conductivity tensor $\boldsymbol{\sigma}$. As we will be interested in oscillatory collective (plasmon) modes, it will be useful to work in terms of Fourier modes, $\rho(\mathbf{r},t) = \rho_\omega(\mathbf{r})e^{i\omega t}$, $\mathbf{j}(\mathbf{r},t) = \mathbf{j}_\omega(\mathbf{r})e^{i\omega t}$, and $\phi(\mathbf{r},t) = \phi_\omega(\mathbf{r})e^{i\omega t}$. The charge and current densities obey a continuity equation, which in terms of the Fourier modes reads:

$$i\omega\rho_\omega(\mathbf{r}) + \nabla\cdot\mathbf{j}_\omega(\mathbf{r}) = 0, \quad \mathbf{j}_\omega(\mathbf{r}) = \boldsymbol{\sigma}(\omega)\nabla\phi_\omega(\mathbf{r}). \quad \text{(S-1)}$$

Importantly, even at $B = 0$, the conductivity $\boldsymbol{\sigma}(\omega)$ possesses off-diagonal contributions, captured by the anomalous Hall conductivity $\sigma_{xy}^{\rm AH}$.

In terms of frequency dependence, the longitudinal conductivity takes a standard Drude form, while the Hall conductivity, arising from the anomalous velocity $\frac{1}{\hbar}\nabla\phi\times\boldsymbol{\Omega}(\mathbf{p})$ [1], is frequency-independent:

$$\sigma_{xx} = \sigma_{yy} = \frac{n_0 e^2/m}{i\tilde{\omega}}, \quad \sigma_{xy}^{\rm AH} = \text{const.} \quad \text{(S-2)}$$

Here $\tilde{\omega} = \omega - i/\tau_{\rm tr}$, where $\tau_{\rm tr}$ is a transport scattering time that arises, e.g., from impurity scattering. We consider collective modes in the regime $\omega\tau_{\rm tr} \gg 1$, where the $1/\tau_{\rm tr}$ contribution to σ_{xx} can be neglected.

The anomalous Hall conductivity is easily connected with quantities in the main text via $\sigma_{xy}^{\rm AH} = \mathcal{F}e^2/h$. Here $\mathcal{F} = \sum_i \int d^2\mathbf{p}\,\Omega_i(\mathbf{p})f_i^0(\mathbf{p})/(2\pi\hbar)^2$ is the dimensionless Berry flux, with $f_i^0(\mathbf{p})$ the equilibrium band occupancy, and $\boldsymbol{\Omega}(\mathbf{p})$ the Berry curvature. Throughout this work we only consider terms up to linear order in $\nabla\phi$.

In this formulation, chiral Berry plasmons (CBPs) at system edges or in disks arise in exactly the same way as shown in the main text, with $\bar{\mathbf{v}}(\mathbf{r},t)$ and $\mathcal{F}$ traded for $\mathbf{j}(\mathbf{r},t)$ and $\sigma_{xy}^{\rm AH}$. That is, Eqs. (S-1) and (S-2) of this supplement, with $1/\tau_{\rm tr} \to 0$, are equivalent to Eqs. (1) and (2) of the main text; following the same steps of derivation yields Eqs. (4-11) of the main text with the replacement $\mathcal{F} = h\sigma_{xy}^{\rm AH}/e^2$.

We note parenthetically that Eq. (S-2) is different from what would be obtained in the analogous case for magnetoplasmons in a system subjected to an applied magnetic field. While both cases feature a nonzero Hall conductivity σ_{xy}, in the case of magnetoplasmons the applied magnetic field induces a Lorentz force that affects electronic motion by modifying the force balance. This leads to different forms of σ_{xx} and σ_{xy} from those in Eq. (S-2); in particular, σ_{xy} in the presence of a finite magnetic field exhibits an ω dependence [2], in contrast to Eq. (S-2). This yields qualitatively different behavior of collective modes. For example, bulk magnetoplasmons are gapped, whereas bulk plasmons in the presence of a Berry flux giving the same dc-Hall conductivity are gapless (see discussion in main text).

CBP dipole mode in a disk

Here we present a more complete analysis of the CBP dipole modes in a disk, which dominate the optical absorption in metallic disks. These dipole modes can be conveniently described through the center of mass (COM) motion, $\{\mathbf{x}(t)\}$, wherein all internal forces cancel (viz. Newton's third law). Here $\{\cdot\}$ denotes the COM average, with $\{\mathbf{x}(t)\} = \int d^2\mathbf{x}\, n(\mathbf{x},t)\mathbf{x}$, and $\{\mathbf{p}(t)\} = \int d^2\mathbf{x}\,\bar{\mathbf{p}}(\mathbf{x},t)$. The COM equations of motion can be obtained from Eq. (1) of the main text via integration by parts, along with the condition that the velocity vanishes when $|\mathbf{x}| \to \infty$. This yields

$$\partial_t\{\mathbf{x}\} = \left\{\frac{\partial\varepsilon}{\partial\mathbf{p}}\right\} + \frac{1}{\hbar}\{e\nabla\phi\times\boldsymbol{\Omega}(\mathbf{p})\}, \quad \partial_t\{\mathbf{p}\} = \{e\nabla\phi\}, \quad \text{(S-3)}$$

where $\mathbf{E}$ is a self-generated electric field associated with the plasmon motion.

To capture the dipolar mode, we use the harmonic potential $-e\phi(\mathbf{x}) = \frac{m}{2}\omega_0^2|\mathbf{x}|^2$ for the electrons (see e.g., Ref. [3]) where ω_0 is the bare plasmon frequency in a disk (with diameter d) in the *absence* of Berry curvature.

In analyzing the COM equations of motion, we first note that $e\nabla\phi(\mathbf{x})$ depends only on $\mathbf{x}$, and $\Omega(\mathbf{p})$ only on $\mathbf{p}$. Keeping terms to linear order in δn, we find the anomalous velocity contribution $\{\mathbf{v}_a\}$ for the COM as

$$\{e\nabla\phi\times\boldsymbol{\Omega}\} = \sum_{\mathbf{x}}\Big[e\nabla\phi(\mathbf{x})\times\sum_i\int\frac{d^2\mathbf{p}}{(2\pi\hbar)^2}\boldsymbol{\Omega}(\mathbf{p})f_i(\mathbf{x},\mathbf{p},t)\Big] = \Big[\sum_{\mathbf{x}} e\nabla\phi(\mathbf{x})\mathcal{F}(\mathbf{x},t)\Big]\times\hat{\mathbf{z}} \approx \zeta\{\mathbf{x}(t)\}\times\hat{\mathbf{z}}, \quad \text{(S-4)}$$

where the Berry flux is given by $\mathcal{F}(\mathbf{x},t) \equiv \sum_i \int d^2\mathbf{p}\,\Omega_i(\mathbf{p}) f_i(\mathbf{x},\mathbf{p},t)/(2\pi\hbar)^2$, the constant ζ can be obtained as detailed below, and i is a band index. Here we will concentrate on the motion of bulk electrons in a given band, n. We therefore take $f_{i<n} = 1$ everywhere inside the disk for bands below n (but vanishing outside the disk). For the purpose of estimating parameters, we make an assumption of local equilibrium and set $f_n(\mathbf{x},\mathbf{p},t) = [e^{\beta(\varepsilon^n_{\mathbf{p}} - \mu(\mathbf{x},t))} + 1]^{-1}$, where $\mu(\mathbf{x},t)$ is a space and time varying chemical potential. Adopting a simple model of a rigidly moving disk of charge with constant density n_0, and density equal to zero outside, gives $\mathcal{F} = \mathcal{F} n(\mathbf{x},t)/n_0$ and $\zeta = -\frac{\mathcal{F}}{n_0} m\omega_0^2$. Here $\mathcal{F}$ is obtained with a fixed uniform chemical potential μ.

Using $\{e\nabla\phi\} = -m\omega_0^2\{\mathbf{x}(t)\}$, Eq. (S-4), and substituting into Eq. (S-3), we obtain the equations of motion for the dipole mode: $(\partial_t^2 + A_{ij})\{x_j\} = 0$, where

$$\mathbf{A} = \begin{pmatrix} \omega_0^2 & \omega_a\partial_t \\ -\omega_a\partial_t & \omega_0^2 \end{pmatrix}, \quad \omega_a = \frac{\mathcal{F}\omega_0^2 m}{n_0\hbar}. \tag{S-5}$$

Here we have used $\left\{\frac{\partial\varepsilon}{\partial\mathbf{p}}\right\} = \frac{1}{m}\{\mathbf{p}\} + \mathcal{O}(\delta n^2)$ and kept only terms up to linear order in δn.

Writing $\{\mathbf{x}\} = \mathbf{x}_0 e^{i\omega t}$, we obtain a secular equation $\mathbf{M}\mathbf{x}_0 = 0$ where $\mathbf{M} = -\omega^2 + \mathbf{A}$, with ∂_t replaced by $i\omega$ within $\mathbf{A}$. Plasmons are given by the zero modes, $\det(\mathbf{M}) = 0$, yielding the split dispersion relation

$$\omega_\pm = \sqrt{\omega_0^2 + \frac{\omega_a^2}{4}} \pm \frac{\omega_a}{2}, \tag{S-6}$$

where $\omega_\pm$, $\omega_0 > 0$. (In the following we will use only positive frequencies; a similar analysis yields the same results for negative $\omega_\pm, \omega_0$ as well). These plasmon modes are *chiral* as they correspond to a rotating COM displacement and momentum

$$\{\mathbf{x}(t)\}_\pm = \frac{|\mathbf{x}_0|}{\sqrt{2}} \begin{pmatrix} 1 \\ \pm i \end{pmatrix} e^{i\omega_\pm t}, \ \{\mathbf{p}(t)\}_\pm = \frac{i\omega_0^2 m}{\omega_\pm}\{\mathbf{x}(t)\}, \tag{S-7}$$

where $\{\mathbf{p}(t)\}_\pm$ and $\{\mathbf{x}(t)\}_\pm$ are offset by a phase of $\pi/2$. As a result, a rotating $\{\mathbf{x}(t)\}_\pm$ gives rise to a circulating momentum/current density (see Fig. 3a of the main text). The clockwise and anticlockwise (ω_+ and ω_-) motions of chiral plasmons sketched in Fig. 3a are oriented for a positive Berry flux, $\mathcal{F}$, pointing in the $\hat{\mathbf{z}}$ direction. This direction sets the orientation of ω_a. For opposite sign of $\mathcal{F}$ the orientations are switched. The distinct frequencies $\omega_\pm$ arise from the anomalous velocity $\{\mathbf{v}_a\}$ adding/subtracting propagation speed from the modes at $\mathcal{F} = 0$.

The CBP dipole mode can manifest in distinct split peaks for optical absorption. This splitting can be analyzed by writing the current density as $\mathbf{j} = en_0\partial_t\{\mathbf{x}\}$ by inverting $\mathbf{M}$ above, and relating current to electric field via $\mathbf{j} = \mathbf{g}\mathbf{E}$, where $\mathbf{E}$ is probing field, and $\mathbf{g}$ is the conductivity tensor. Optical absorption is characterized by the real part of the longitudinal conductivity, $\mathbf{g}_{\rm xx}$, as

$$\mathrm{Re}[g_{\rm xx}(\omega)] = \frac{1}{2}\sum_\pm \frac{\mathcal{D}\Gamma\omega^2}{(\omega^2 \pm \omega\omega_a - \omega_0^2)^2 + \Gamma^2\omega^2}, \tag{S-8}$$

where $\mathcal{D} = \frac{n_0 e^2}{m}$ is the Drude weight, and Γ is the transport relaxation rate, included phenomenologically via $\partial_t^2 \to \partial_t^2 + \Gamma\partial_t$. This yields the split peak optical absorption for the disk geometry shown in Fig. 3b of the main text.

Optical Selection rules for gapped Dirac materials

Here we detail how circularly polarized light can yield valley selectivity in gapped Dirac materials (GDMs) where inversion symmetry has been broken. This selectivity arises due to the pseudo-spinor nature of the wavefunctions on the A/B sublattices. The selection rules were derived in Ref. [4] by analyzing the orbital magnetic moments of electronic wavepackets in the K and K' valleys. For the reader's convenience, here we present an alternative calculation based on Fermi's golden rule.

The low energy Hamiltonian for gapped Dirac materials can be described as $\mathcal{H} = H_K + H_{K'}$, where

$$H_K = v\boldsymbol{\sigma}_+ \cdot \mathbf{p}_K + \Delta\sigma_z, \quad H_{K'} = v\boldsymbol{\sigma}_- \cdot \mathbf{p}_{K'} + \Delta\sigma_z, \tag{S-9}$$

where $H_K, H_{K'}$ describe electrons close to the K and K' points, v is the Fermi velocity, and $\boldsymbol{\sigma}_\pm = \sigma_x\hat{\mathbf{x}} \pm \sigma_y\hat{\mathbf{y}}$, with $\sigma_{x,y,z}$ the Pauli matrices. Here $\mathbf{p}_{K,K'}$ describe momenta taken relative to the K and the K' points, respectively, and 2Δ is the gap size. In the following we shall drop the explicit K and K' labels on $\mathbf{p}_{K,K'}$ for brevity.

A variety of systems obey Eq. (S-9), including van der Waals heterostructures where A/B sublattice symmetry has been broken as in G/hBN heterostructures. In such systems the gap size 2Δ corresponds to the asymmetry of the potential on the A/B sublattices. Below we will focus on this case for concreteness. However, the underlying physics is general and applies to a broad range of GDMs, such as dual-gated bilayer graphene and transition metal dichalcogenides, where inversion symmetry has been broken.

The eigenfunctions of Eq. (S-9) can be expressed as pseudo-spinors

$$|+\rangle_{K(K')} = \begin{pmatrix} \cos\frac{\theta}{2} e^{-i\tau\phi} \\ \sin\frac{\theta}{2} \end{pmatrix}, \quad |-\rangle_{K(K')} = \begin{pmatrix} \sin\frac{\theta}{2} e^{-i\tau\phi} \\ -\cos\frac{\theta}{2} \end{pmatrix}, \tag{S-10}$$

where $|\pm\rangle$ denote states in the conduction (valence) band, $\tau = 1$ for valley K and $\tau = -1$ for valley K', $\tan\theta = v|\mathbf{p}|/\Delta$, and $\tan\phi = p_y/p_x$. Here the energy eigenvalues are $\varepsilon^\pm_{\mathbf{p}} = \pm\sqrt{v^2|\mathbf{p}|^2 + \Delta^2}$.

We proceed by noting that when light with frequency $\hbar\omega \geq 2\Delta$ is incident on the GDM, electrons in the valence band can be excited into the conduction band. The light-matter coupling is captured by writing $\mathbf{p} \to \mathbf{p} - e\mathbf{A}/c$, where the vector potential $\mathbf{A}$ is related to the incident light electric field $\mathbf{E}$ via $\mathbf{A} = \frac{ic}{\omega}\mathbf{E}$. Here $\mathbf{E} = \mathbf{E}_0 e^{i\omega t}$ for light with frequency ω. The rate of electron-hole pair creation (absorption of photons) can be calculated via Fermi's golden rule

$$W_{K(K')} = \frac{2\pi}{\hbar} \sum_{\mathbf{p}} |M_{\mathbf{p}}^{K(K')}|^2 \delta(\varepsilon_{\mathbf{p}}^{+} - \hbar\omega/2), \qquad \text{(S-11)}$$

with the matrix elements

$$\begin{aligned} M_{\mathbf{k}}^{K} &= \frac{iev}{\omega} \langle + | E_x \sigma_x \; + \; E_y \sigma_y | - \rangle_K \\ M_{\mathbf{k}}^{K'} &= \frac{iev}{\omega} \langle + | E_x \sigma_x \; - \; E_y \sigma_y | - \rangle_{K'}. \end{aligned} \qquad \text{(S-12)}$$

We note that the different signs in front of E_y in $M_{\mathbf{k}}^{K}$ and $M_{\mathbf{k}}^{K'}$ arise from the different way $\mathbf{A}$ couples to pseudospin in H_K and $H_{K'}$ (see Eq. S-9).

With the help of the identities

$$\begin{aligned} \langle + | \sigma_x | - \rangle_{K(K')} &= \sin^2\tfrac{\theta}{2} e^{-i\tau\phi} - \cos^2\tfrac{\theta}{2} e^{i\tau\phi} \\ \langle + | \sigma_y | - \rangle_{K(K')} &= i\big(\sin^2\tfrac{\theta}{2} e^{-i\tau\phi} + \cos^2\tfrac{\theta}{2} e^{i\tau\phi}\big), \end{aligned} \qquad \text{(S-13)}$$

and writing $\mathbf{E}_0 = |\mathbf{E}_0| \, (\hat{\mathbf{x}} + i\eta\hat{\mathbf{y}})/\sqrt{2}$ for left-handed (LH, $\eta = 1$) and right-handed (RH, $\eta = -1$) circularly polarized light, we obtain

$$W_K^{\eta} = W_0 \left[\frac{2\Delta}{\hbar\omega} + \eta \right]^2, \quad W_{K'}^{\eta} = W_0 \left[\frac{2\Delta}{\hbar\omega} - \eta \right]^2. \qquad \text{(S-14)}$$

Here $W_0 = N_s e^2 |\mathbf{E}_0|^2/(16\hbar^2\omega)$, where N_s is the spin degeneracy.

Equation (S-14) shows that electron-hole transitions in the valleys K/K' can be selectively excited using LH/RH circularly polarized light (see Fig. 3c inset of the main text). Indeed for $\hbar\omega = 2\Delta$, perfect selection of K or K' electron-hole transitions can be achieved, in agreement with Ref. [4]. We emphasize that this selectivity comes from the *orbital* physics of light-matter coupling; it does not require or involve spin-orbit coupling and can even arise in materials with negligible spin orbit-coupling as modeled by Eq. (S-9).

Optically pumped "On-Demand" CBP dipole mode in gapped Dirac materials

Here we consider how CBPs might arise in non-magnetic gapped Dirac materials such as MoS_2, or gapped G/h-BN. To analyze this, we focus on nominally time reversally invariant gapped Dirac materials. We model the valley dependent $\mathcal{F}$ as [5]: $\mathcal{F}_{K,K'} = \tau_z N_s \frac{\tilde{n}^{1/2}}{2(n_{K,K'} + \tilde{n})^{1/2}} \operatorname{sgn}\Delta$, where $n_{K,K'}$ are the valley carrier densities, $\tau_z = \mp$ for the K, K' valleys, $N_s = 2$ is the spin degeneracy, and $\tilde{n} = \Delta^2/4\pi v_F^2 \hbar^2$ gives a characteristic density scale. The bandgap is $2|\Delta|$. When $n_K = n_{K'}$, the total flux $\mathcal{F} = \mathcal{F}_K + \mathcal{F}_K'$ vanishes.

However, a non-vanishing net Berry flux $\mathcal{F}_{\rm pe}$ for the photoexcited system is achieved when $n_K \neq n_{K'}$ (Fig. 3c inset of the main text). Setting $n_{K'} = 0$, giving $\mathcal{F}_{K'} = +1$, the valley polarized electron ($n_{\rm el}$) and hole ($n_{\rm h}$) populations concentrated at the K valley conduction and valence band extrema yield

$$\begin{aligned} \mathcal{F}_{\rm pe} &\approx 1 - \frac{1}{2} \Big[\frac{\tilde{n}^{1/2}}{\sqrt{\tilde{n} + n_{\rm h}}} + \frac{\tilde{n}^{1/2}}{\sqrt{\tilde{n} + n_{\rm el}}} \Big], \\ \tilde{n} &= 1.8 \times 10^{13} \frac{(\Delta[\mathrm{eV}])^2}{(v_F[\mathrm{cm\,s^{-1}}]/10^8)^2} \mathrm{cm}^{-2}. \end{aligned} \qquad \text{(S-15)}$$

Here we have used that Berry curvature (including its sign) is the same for electrons and holes, following from particle-hole symmetry of the Dirac Hamiltonian, and used the convention $n_{\rm el,h} > 0$. In deriving Eq. (S-15), we used the spin degeneracy $N_s = 2$ for the K' valley, and noted that only a single spin species in the K valley is excited by the circularly polarized light.

For demonstration we examine CBPs in the disk geometry as above. We analyze the coupled motion of photoexcited electrons and holes in a single valley via COM coordinates ($\{\mathbf{x}_{\rm el}\}$) and ($\{\mathbf{x}_{\rm h}\}$), respectively, giving:

$$\left[\partial_t^2 + \begin{pmatrix} \mathbf{A}_{\rm el} & -\mathbf{A}_{\rm el} \\ -\mathbf{A}_{\rm h} & \mathbf{A}_{\rm h} \end{pmatrix} \right] \begin{pmatrix} \{\mathbf{x}_{\rm el}\} \\ \{\mathbf{x}_{\rm h}\} \end{pmatrix} = 0, \qquad \text{(S-16)}$$

where $\mathbf{A}_{\rm el}$ and $\mathbf{A}_{\rm h}$ are defined as in Eq. (7) of the main text, with the density n_0 and plasmon mass m replaced by the appropriate values for electrons or holes. In writing these equations, we have used that the restoring force arising from the mutual attraction of photoexcited electron and hole subsystems is $\{e\nabla\phi\}_{\rm el(h)} \approx \mp\alpha(\{\mathbf{x}_{\rm el}\} - \{\mathbf{x}_{\rm h}\})$, where $\{\cdot\}_{\rm el,h}$ denote the COM averages for electron and hole distributions and the upper (lower) sign is for electrons (holes). Here α characterizes the strength of the electron-hole interaction. For brevity, in the following analysis we set $\mathbf{A}_{\rm el} = \mathbf{A}_{\rm h}$, giving $\alpha = m\omega_0^2$, where ω_0 is the plasmon frequency associated with a unipolar system with carrier density $n = n_{\rm el} = n_{\rm h}$.

Zero modes of Eq. (S-16) with $\partial_t^2 \to -\omega^2$ yield CBPs with $\{\mathbf{x}_{\rm el}\} = -\{\mathbf{x}_{\rm h}\}$, giving:

$$\omega_{\pm}^{\rm el-h} = \sqrt{\omega_a^2 + 2\omega_0^2} \pm \omega_a, \qquad \text{(S-17)}$$

where ω_a is given by Eq. (7) of the main text with $\mathcal{F}$ replaced by $\mathcal{F}_{\rm pe}$. Importantly, $\omega_{\pm}^{\rm el-h}$ chiral (electron-hole) plasmons feature *co-rotating* $\{\mathbf{x}_{\rm el}\}$ and $\{\mathbf{x}_{\rm h}\}$. This dipole like rotation follows from the equal sign of $\mathcal{F}$ for electrons and holes. Modes with $\{\mathbf{x}_{\rm el}\} = \{\mathbf{x}_{\rm h}\}$ are also eigenmodes of Eq. (S-16). However, they have frequency, $\omega = 0$.

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