Supplemental material: Chiral plasmon in gapped Dirac systems

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I. Von Neumann equation for density matrix

Evolution of electron subsystem in massive Dirac system (MDS) in external electromagnetic field \( \mathbf{E} \) can be described using the von Neumann equation for electron statistical operator

\[
 i\hbar \frac{\partial \hat{\rho}}{\partial t} = [\mathcal{H} + \mathbf{V}, \hat{\rho}] \tag{1}
\]

where \( \mathcal{H} \) is the single-electron Hamiltonian of MDS, \( \mathbf{V} = -e \mathbf{E} \cdot \mathbf{r} \) is the interaction term, and \( e = -1.6 \times 10^{-19} \) is the electron charge. In the basis of eigenfunctions of \( \mathcal{H} \) Eq. (1) has the form

\[
 i\hbar \frac{\partial \rho_{kj,k'j'}}{\partial t} = [\mathcal{H}, \hat{\rho}]_{kj,k'j'} + [\mathbf{V}, \hat{\rho}]_{kj,k'j'}, \tag{2}
\]

where

\[
 A_{kj,k'j'} = \int_S \Psi_{kj}(\mathbf{r}) \hat{A} \Psi_{k'j'}(\mathbf{r}) d^2 \mathbf{r}, \tag{3}
\]

\( \hat{A} = \hat{\rho}, \mathcal{H}, \mathbf{V}, \mathbf{k} \) is two-dimensional electron wavevector, \( j \) is an index designating quantum number of electrons in MDS. Integration is taken over the surface area \( S \) of MDS. Functions \( \Psi_{kj}(\mathbf{r}) \) are solutions of the equation

\[
 \mathcal{H} \Psi_{kj}(\mathbf{r}) = \varepsilon_{kj} \Psi_{kj}(\mathbf{r}) \tag{4}
\]

and can be written in the Bloch form

\[
 \Psi_{kj}(\mathbf{r}) = \frac{1}{\sqrt{N}} e^{i \mathbf{k} \cdot \mathbf{r}} u_{kj}(\mathbf{r}), \tag{5}
\]

where \( N \) is the number of unit cells in MDS, and \( u_{kj}(\mathbf{r}) = u_{kj}(\mathbf{r} + \mathbf{R}_n) \) is Bloch amplitude, \( \mathbf{R}_n \) is the vector of MDS lattice.

First term on right-hand side of Eq. (2) can be written as

\[
 [\mathcal{H}, \hat{\rho}]_{kj,k'j'} = \rho_{kj,k'j'} (\varepsilon_{kj} - \varepsilon_{k'j'}), \tag{6}
\]

while second term takes form

\[
 [\mathbf{V}, \hat{\rho}]_{kj,k'j'} = \sum_{\mathbf{k}''j''} (V_{kj,k''j''} \rho_{k''j'',k'j'} - \rho_{kj,k''j''} V_{k''j'',k'j'}). \tag{7}
\]

Let us consider first term on the left hand side of Eq. (7),

\[
 \sum_{\mathbf{k}''j''} V_{kj,k''j''} \rho_{k''j'',k'j'} = -e \mathbf{E} \cdot \sum_{\mathbf{k}''j''} \rho_{k''j'',k'j'} \int_S \Psi_{kj}(\mathbf{r}) \Psi_{k''j''}(\mathbf{r}) d^2 \mathbf{r}
\]

\[
 = \frac{ie}{\sqrt{N}} \mathbf{E} \cdot \sum_{\mathbf{k}''j''} \rho_{k''j'',k'j'} \int_S \left[ -\frac{\partial u_{kj}^*(\mathbf{r}) e^{-i \mathbf{k} \cdot \mathbf{r}}}{\partial \mathbf{k}} \Psi_{k''j''}(\mathbf{r}) + \frac{\partial u_{kj}^*(\mathbf{r})}{\partial \mathbf{k}} e^{-i \mathbf{k} \cdot \mathbf{r}} \Psi_{k''j''}(\mathbf{r}) \right] d^2 \mathbf{r} \tag{8}
\]

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As electron wavefunctions are normalized,

\[ \int_S \Psi_{k_j}^*(r) \Psi_{k'_{j'}}(r) \, d^2r = \delta_{kk'} \delta_{jj'}, \]  

(9)

the first term in square brackets in Eq. (8) takes form

\[ \frac{1}{\sqrt{N}} \sum_{k''_{j''}} \rho_{k''_{j''}, k_{j'}} \int_S \frac{\partial u_{k_j}^*(r)}{\partial k} e^{-i \mathbf{k} \cdot \mathbf{r}} \Psi_{k''_{j''}}(r) \, d^2r = \frac{\partial}{\partial \mathbf{k}} \sum_{k''_{j''}} \rho_{k''_{j''}, k_{j'}} \int_S \Psi_{k_j}^*(r) \Psi_{k''_{j''}}(r) \, d^2r = \frac{\partial \rho_{k_j, k'_{j'}}}{\partial \mathbf{k}}. \]  

(10)

In order to simplify the second term in the square brackets we split integral over the surface area \( S \) as sum of integrals over unit cells \( \Omega_n \):

\[ \frac{1}{N} \int_S \frac{\partial u_{k_j}^*(r)}{\partial k} \left( u_{k''_{j''}}(r + \mathbf{R}_n) e^{i(\mathbf{k''} - \mathbf{k}) \cdot \mathbf{r}} \right) \, d^2r = \frac{1}{N} \sum_{n=1}^N \int_{\Omega_n} \frac{\partial u_{k_j}^*(r)}{\partial k} u_{k''_{j''}}(r + \mathbf{R}_n) e^{i(\mathbf{k''} - \mathbf{k}) \cdot (\mathbf{r} + \mathbf{R}_n)} \, d^2r \]

\[ = \frac{1}{N} \sum_{n=1}^N e^{i(\mathbf{k''} - \mathbf{k}) \cdot \mathbf{R}_n} \int_{\Omega_1} \frac{\partial u_{k_j}^*(r)}{\partial k} u_{k''_{j''}}(r) e^{i(\mathbf{k''} - \mathbf{k}) \cdot \mathbf{r}} \, d^2r \]

\[ = \delta_{kk''} \int_{\Omega_1} \frac{\partial u_{k_j}^*(r)}{\partial k} u_{k''_{j''}}(r) \, d^2r = -\delta_{kk''} \int_{\Omega_1} u_{k_j}^*(r) \frac{\partial u_{k''_{j''}}(r)}{\partial k} \, d^2r. \]  

(11)

In order to obtain last expression we integrated by parts and took into account periodicity of Bloch amplitudes at the unit cell boundaries.

Plugging Eqs. (10), (11) back into Eq. (8) we obtain

\[ (V \hat{\rho})_{k_j, k'_{j'}} = -ie \mathbf{E} \cdot \frac{\partial \rho_{k_j, k'_{j'}}}{\partial \mathbf{k}} - e \mathbf{E} \cdot \sum_{j''} \mathbf{R}_{j''_{j'}}(\mathbf{k}) \rho_{k_{j''}, k'_{j'}}. \]  

(12)

where

\[ \mathbf{R}_{j''_{j'}}(\mathbf{k}) = \frac{i}{2} \int_{\Omega_1} \left( u_{k_j}^*(r) \frac{\partial u_{k_{j''}}(r)}{\partial k} - \frac{\partial u_{k_j}^*(r)}{\partial k} u_{k_{j''}}(r) \right) \, d^2r. \]  

(13)

Repeating the steps above we also obtain

\[ (\hat{\rho} V)_{k_j, k'_{j'}} = ie \mathbf{E} \cdot \frac{\partial \rho_{k_j, k'_{j'}}}{\partial k'} - e \mathbf{E} \cdot \sum_{j''} \rho_{k_j, k'_{j'}} \mathbf{R}_{j''_{j'}}(k'), \]  

(14)

and

\[ [V, \hat{\rho}]_{k_j, k'_{j'}} = -ie \mathbf{E} \cdot \left( \frac{\partial \rho_{k_j, k'_{j'}}}{\partial \mathbf{k}} + \frac{\partial \rho_{k_j, k''_{j'}}}{\partial \mathbf{k'}} \right) - e \mathbf{E} \cdot \sum_{j''} \left( \mathbf{R}_{j''_{j'}}(k) \rho_{k_{j''}, k'_{j'}} - \rho_{k_j, k'_{j'}} \mathbf{R}_{j''_{j'}}(k') \right). \]  

(15)

Thus, the equation of motion for the electron density matrix takes form

\[ i\hbar \frac{\partial \rho_{k_j, k'_{j'}}}{\partial t} = \rho_{k_j, k'_{j'}} (\hat{\mathcal{E}}_k - \hat{\mathcal{E}}_{k'}) - i e \mathbf{E} \cdot \left( \frac{\partial \rho_{k_j, k'_{j'}}}{\partial \mathbf{k}} + \frac{\partial \rho_{k_j, k''_{j'}}}{\partial \mathbf{k'}} \right) \]

\[ - e \mathbf{E} \cdot \sum_{j''} \left( \mathbf{R}_{j''_{j'}}(k) \rho_{k_{j''}, k'_{j'}} - \rho_{k_j, k'_{j'}} \mathbf{R}_{j''_{j'}}(k') \right). \]  

(16)

Eq. (16) accounts both for the intraband motions and direct and indirect interband transitions. However, contribution of electrons indirect interband transitions to the optical response is negligible and thus can be omitted. Then, in the
The non-diagonal matrix elements evolve as
\[
\frac{\partial \rho_{j'j}(t, \mathbf{k})}{\partial t} + \frac{e}{\hbar} \mathbf{E} \cdot \frac{\partial \rho_{j'j}(t, \mathbf{k})}{\partial \mathbf{k}} = \frac{i}{\hbar} \rho_{j'j}(t, \mathbf{k}) [\mathcal{E}_{k_j} - \mathcal{E}_{k_{j'}}] + \frac{i e}{\hbar} \mathbf{E} \cdot \sum_{j''} [\mathbf{R}_{j''j}(\mathbf{k}) \rho_{j''j'}(t, \mathbf{k}) - \rho_{j''j'}(t, \mathbf{k}) \mathbf{R}_{j''j'}(\mathbf{k})],
\]
where \( \rho_{j'j}(t, \mathbf{k}) = \rho_{kj',\nu j'}(t) \), where \( \nu, \mu = c, v \) denote electron and hole bands.

II. Incorporating relaxation into equations of motion for density matrix

Electromagnetic radiation can not also couple electrons in different valleys in \( \mathbf{k} \)-space, and thus it is convenient to write equations separately for each of the valleys. We account for different valleys by introducing valley quantum number \( \tau = \pm 1 \) (or \( K, K' \)). For convenience, we designate density matrix elements as \( \rho_{j'j}(t, \mathbf{k}) = \rho_{\nu \mu j'j}(t, \mathbf{k}) \).

We introduce relaxation phenomenologically using relaxation time approximation. Then the equation for the density matrix elements of electrons in conduction band in \( K \) valley takes form
\[
\frac{\partial \rho_{cc}^{K}(t, \mathbf{k})}{\partial t} + \frac{e}{\hbar} \mathbf{E} \cdot \frac{\partial \rho_{cc}^{K}(t, \mathbf{k})}{\partial \mathbf{k}} = - \frac{\rho_{cc}^{K}(t, \mathbf{k}) - \rho_{cc}^{eq,K}(\mathbf{k})}{\tau_0} + \frac{\rho_{cc}^{K'K}(t, \mathbf{k}) - \rho_{cc}^{K}(t, \mathbf{k})}{\tau_1} + \frac{i e}{\hbar} \mathbf{E} \cdot [\mathbf{R}_{cc}^{K'(K)}(\mathbf{k}) \rho_{cc}^{K}(t, \mathbf{k}) - \rho_{cc}^{K}(t, \mathbf{k}) \mathbf{R}_{cc}^{K'}(\mathbf{k})],
\]
where \( \rho_{cc}^{eq,K}(\mathbf{k}) \) is equilibrium Fermi-Dirac distribution, \( \tau_0 \) is the population relaxation time. We also took into account that electron-phonon interactions can couple electrons in different valleys by introducing inter-valley scattering time \( \tau_1 \). The rest of equations for diagonal matrix elements can be written as
\[
\frac{\partial \rho_{cc}^{K'}(t, \mathbf{k})}{\partial t} + \frac{e}{\hbar} \mathbf{E} \cdot \frac{\partial \rho_{cc}^{K'}(t, \mathbf{k})}{\partial \mathbf{k}} = - \frac{\rho_{cc}^{K'}(t, \mathbf{k}) - \rho_{cc}^{eq,K'}(\mathbf{k})}{\tau_0} + \frac{\rho_{cc}^{K''K'}(t, \mathbf{k}) - \rho_{cc}^{K'}(t, \mathbf{k})}{\tau_1} + \frac{i e}{\hbar} \mathbf{E} \cdot [\mathbf{R}_{cc}^{K''K'}(\mathbf{k}) \rho_{cc}^{K'}(t, \mathbf{k}) - \rho_{cc}^{K'}(t, \mathbf{k}) \mathbf{R}_{cc}^{K''}(\mathbf{k})],
\]
\[
\frac{\partial \rho_{cc}^{K''}(t, \mathbf{k})}{\partial t} + \frac{e}{\hbar} \mathbf{E} \cdot \frac{\partial \rho_{cc}^{K''}(t, \mathbf{k})}{\partial \mathbf{k}} = - \frac{\rho_{cc}^{K''}(t, \mathbf{k}) - \rho_{cc}^{eq,K''}(\mathbf{k})}{\tau_0} + \frac{\rho_{cc}^{K''K''}(t, \mathbf{k}) - \rho_{cc}^{K''}(t, \mathbf{k})}{\tau_1} + \frac{i e}{\hbar} \mathbf{E} \cdot [\mathbf{R}_{cc}^{K''K''}(\mathbf{k}) \rho_{cc}^{K''}(t, \mathbf{k}) - \rho_{cc}^{K''}(t, \mathbf{k}) \mathbf{R}_{cc}^{K''}(\mathbf{k})].
\]

The non-diagonal matrix elements evolve as
\[
\frac{\partial \rho_{cc}^{K'}(t, \mathbf{k})}{\partial t} + \frac{e}{\hbar} \mathbf{E} \cdot \frac{\partial \rho_{cc}^{K'}(t, \mathbf{k})}{\partial \mathbf{k}} = \left( i \omega_{cc}^{K} + \frac{1}{\tau_d} \right) \rho^{K}_{cc} + \frac{2i e}{\hbar} \mathbf{E} \cdot \mathbf{R}_{cc}^{K'} \rho^{K}_{cc} - \frac{i e}{\hbar} \mathbf{E} \cdot \mathbf{R}_{cc}^{K} \rho^{K}_{in},
\]
where \( \omega_{cc}^{K} = \omega_{cc}^{K} - \omega_{cc}^{K} \), \( \omega_{cc}^{K} = (\mathcal{E}_{cc}^{K} - \mathcal{E}_{cc}^{K}) / \hbar \), and \( \tau_d \) is electron dephasing time.

III. Eigenenergies and eigenfunctions

In order to calculate dipole matrix elements we need to know Bloch amplitudes \( u_{kj} \). We use MDS Hamiltonian
\[
\mathcal{H} = \frac{\Delta}{2} \mathbf{\sigma}_z + t_0 a_0 \mathbf{k} \cdot \mathbf{\sigma},
\]
where \( \mathbf{\sigma}_\tau = (\mathbf{\tau}_{\mathbf{\sigma}_x}, \mathbf{\sigma}_y) \). After diagonalizing the Hamiltonian, we obtain eigenenergies
\[
\mathcal{E}_{k\nu}^{\tau} = \frac{\nu}{2} \sqrt{\mathcal{E}_{k\nu}^{\tau}},
\]
and eigenfunctions
\[
u_{k\nu}^{\tau} = \frac{1}{\sqrt{S_{\Omega}}} \Phi_k^{\tau} \left( \begin{array}{c} 1 \\ W_k^{\tau} \end{array} \right), \quad u_{k\nu}^{\tau} = \frac{1}{\sqrt{S_{\Omega}}} \Phi_k^{\tau} \left( \begin{array}{c} -W_k^{\tau*} \\ 1 \end{array} \right),
\]
In the case $k_K$ Thus (+) polarized wave pumps $K$ valley. Let us consider $\nu = \pm 1$ denotes conduction and valence band respectively, and $S_0$ is an area of unit cell.

IV. Dipole matrix elements $R_{jj'}$

$$W_K = \frac{2t_0a_0(\tau k_x + ik_y)}{\Delta + \sqrt{\beta_k^2}}, \quad |\Phi_k'|^2 = 1 + |W_K|^2,$$  \hfill (25)

$$\beta_K^2 = 4a_0^2\alpha^2 k^2 + \Delta^2,$$  \hfill (26)

$$\nu = \pm 1$$

Dot product $E_0 \cdot R^{K,K'}_{jj'}$ defines valley interaction with a field of a given polarization. Let us assume that $E_0 = |E_0|(e_x + i e_y)$. Then for $K$ valley ($\tau_z = 1$)

$$E_0 \cdot R^{K}_{jj'} = |E_0| \frac{a_0 t}{\sqrt{\beta_k^2 \alpha_k}} (-2i\alpha_k^K + 4ia_0^2 t^2 (k_x^2 + k_y^2))$$  \hfill (32)

In the case $k_x = k_y = 0$

$$E_0 \cdot R^{K}_{jj'} = -2i|E_0| \frac{a_0 t}{\sqrt{\beta_k^2}}$$  \hfill (33)

Thus (+) polarized wave pumps $K$ valley. Let us consider $K'$ valley ($\tau_z = -1$) now:

$$E_0 \cdot R^{K'}_{jj'} = |E_0| \frac{4ia_0^2 t^3 (k_x^2 - k_y^2)}{\sqrt{\beta_k^2 \alpha_k}}$$  \hfill (34)

In the case $k_x = k_y = 0$

$$E_0 \cdot R^{K'}_{jj'} = 0.$$  \hfill (35)

Thus electrons in $K'$ valley do not interact with (+) polarized wave.
V. MDS interaction with laser pump

Let us consider interaction of MDS with monochromatic electromagnetic wave

\[ E = E_0 e^{-i\omega t} + c.c. \]  \quad (36)

We assume that frequency of the wave \( \omega \) is in the vicinity of the frequency of electron interband transitions. Eqs. (18)-(22) are partial differential equations which have to be solved numerically in the general case. However, in the case of near-resonant wave interband electron transitions dominate intraband motion. Thus we can neglect terms containing partial derivatives over \( k \) on the right-hand side.

Under the influence of electromagnetic field non-diagonal matrix elements evolve as \( \rho_{cv} = \hat{\rho}_{cv}(t)e^{-i\omega t} \), where \( \hat{\rho}_{cv}(t) \) is a slow varying amplitude. We are interested in the steady-state solution. Using the rotation-wave approximation and setting partial derivative of \( \hat{\rho}_{cv}(t) \) to zero, we obtain

\[ \hat{\rho}_{cv} = \frac{e}{\hbar(\omega - \omega_{cv}^0 + i/\tau_d)} E_0 \cdot \mathbf{R}_{cv}^T \hat{\rho}_{in}. \]  \quad (37)

Eqs. (18)-(21) then take form

\[ 0 = -\frac{\rho^K_{cc} - \rho_{cc},K}{\tau_0} + \frac{\rho^K_{cc} - \rho_{cc}'}{\tau_1} + \frac{i e}{\hbar} [E_0 \cdot \mathbf{R}_{cc}^T \rho^K_{cc} - E_0^* \cdot \mathbf{R}_{cc}^T \rho^K_{cc}'], \]  \quad (38)

\[ 0 = -\frac{\rho^K_{cc} - \rho_{cc},K'}{\tau_0} + \frac{\rho^K_{cc} - \rho_{cc}''}{\tau_1} + \frac{i e}{\hbar} [E_0 \cdot \mathbf{R}_{cc}^T \rho^K_{cc}'' - E_0^* \cdot \mathbf{R}_{cc}^T \rho^K_{cc}']. \]  \quad (39)

\[ 0 = -\frac{\rho^K_{cc} - \rho_{cc},K'}{\tau_0} + \frac{\rho^K_{cc} - \rho_{cc}''}{\tau_1} - \frac{i e}{\hbar} [E_0 \cdot \mathbf{R}_{cc}^T \rho^K_{cc}'' - E_0^* \cdot \mathbf{R}_{cc}^T \rho^K_{cc}'], \]  \quad (40)

\[ 0 = -\frac{\rho^K_{cc} - \rho_{cc},K'}{\tau_0} + \frac{\rho^K_{cc} - \rho_{cc}''}{\tau_1} - \frac{i e}{\hbar} [E_0 \cdot \mathbf{R}_{cc}^T \rho^K_{cc}'' - E_0^* \cdot \mathbf{R}_{cc}^T \rho^K_{cc}']. \]  \quad (41)

By substituting Eq. (37) into equations (38)-(41), we obtain system of four linear equations for diagonal matrix elements in two valleys

\[ \hat{\rho}_{vv}^K = \beta_K \left( \rho_{vv}^K + \gamma \rho_{vv}^{K'} + \alpha_K \rho_{cc}^{K} \right), \]

\[ \hat{\rho}_{vv'}^{K'} = \beta_{K'} \left( \rho_{vv'}^{K'} + \gamma \rho_{vv}^{K} + \alpha_K \rho_{cc}^{K'} \right), \]

\[ \hat{\rho}_{cc}^K = \beta_K \left( \rho_{cc}^K + \gamma \rho_{cc}^{K'} + \alpha_K \rho_{vv}^{K} \right), \]

\[ \hat{\rho}_{cc'}^{K'} = \beta_{K'} \left( \rho_{cc'}^{K'} + \gamma \rho_{cc}^{K} + \alpha_K \rho_{vv'}^{K'} \right), \]

where

\[ \beta_K = \frac{1}{1 + \alpha_K + \gamma}, \quad \gamma = \frac{\tau_0}{\tau_1}, \]

\[ \alpha_{K,K'} = \frac{2e^2\tau_0\tau_d|E_0 \cdot \mathbf{R}_{cc}^{K,K'}|^2}{\hbar^2 \left( \tau_d^2 (\omega - \omega_{cv}^{K,K'})^2 + 1 \right)}. \]

In the paper we assumed that \( \tau_d = \tau_0 \).