Topological properties of Weyl semimetals in circularly-polarized ultrafast laser field

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We study the topological properties of three-dimensional Weyl semimetals in an ultrafast laser pulse. A single oscillation of circularly-polarized pulse induces the finite conduction band population in the reciprocal space which is highly structured and is determined by the topological phase. These textures can be probed by a linear polarized pulse which is applied after the circularly-polarized pulse. The response of the system to the linear probe pulse is highly dependent on the chirality of the circularly-polarized pulse. Also, we show that the induction conduction band population by a circularly-polarized pulse consisting of two oscillations with opposite chirality in the reciprocal space is highly chiral which represent the intrinsic chirality of the Weyl nodes.

I. INTRODUCTION

Weyl semimetals (WS) are topological materials characterized by linear band crossing points called Weyl points near the Fermi level. Weyl points appear in pairs and in crystals either time reversal symmetry $\mathcal{T}$ or inversion symmetry $\mathcal{P}$ is broken, the Weyl nodes of opposite chirality are separated in momentum space and they are connected only through the Fermi arcs [1]. The surface Fermi arcs have been observed in such materials by ARPES technique [2]. Weyl semimetals pose remarkable properties both in bulk and on the surface [3].

Because of the strong spin-orbit coupling (SOC), the Weyl points act as a sink or source of Berry curvature in the Brillouin zone. The physical consequences of Berry curvature in Weyl semimetals has been discussed in the context of interesting phenomena such as anomalous Hall effect when the time reversal symmetry (TRS) is broken in the system [4], dc photocurrent induced in Weyl semimetals as the nonlinear optical response [5] and quantum nonlinear Hall effect in time-reversal invariant Weyl semimetals [6]. Ref. [7] reported how to detect the chirality of Weyl fermions by measuring the photocurrent in response to circularly polarized mid-infrared light. Also, it has been shown that the chirality of a particular Weyl point can be determined by circular dichroism (CD) of time-dependent ARPES experiment [9].

In this paper, we consider another consequence of Berry curvature dipole in such materials. We theoretically study the interaction of Weyl semimetals with an ultrafast laser pulse. We show that a circularly-polarized ultrafast pulse can excite and control Weyl points selectively which manifest itself in a texture of the electron conduction band (CB) population in momentum space. The texture is highly structured and is determined by the topological phase which is a consequence of Berry curvature in Weyl semimetals has been discussed in the context of interesting phenomena such as anomalous Hall effect when the time reversal symmetry (TRS) is broken in the system [4], dc photocurrent induced in Weyl semimetals as the nonlinear optical response [5] and quantum nonlinear Hall effect in time-reversal invariant Weyl semimetals [6].

II. MODEL AND EQUATIONS

The Hamiltonian of an electron in Weyl semimetals in an external electric field reads as

$$\mathcal{H} = \mathcal{H}_0 + e \mathbf{F}(t) \mathbf{r}, \quad (1)$$

where $\mathcal{H}_0$ is a field-free electron Hamiltonian, $e$ is electron charge and $\mathbf{F}(t)$ is the vector of the field, and $\mathbf{r}$ is the vector of position. We assume a two band Bloch Hamiltonian for Weyl semimetals in three dimensions as [9]

$$\mathcal{H}_0 = A(k) \sigma_x + B(k) \sigma_y + C(k) \sigma_z, \quad (2)$$

$$A(k) = t_x (\cos(k_x a) - \cos(k_0 a)) + t_y (\cos(k_y b) - 1) + t_z (\cos(k_z c) - 1),$$

$$B(k) = t_y \sin(k_y b),$$

$$C(k) = t_z \sin(k_z c),$$

where $a$, $b$ and $c$ are lattice constants in $x$, $y$ and $z$ directions, respectively, see Fig.1(a). $t_x$, $t_y$ and $t_z$ are hopping parameters, $k^\pm_0 = (\pm k_0, 0, 0)$, is the position of Weyl nodes in momentum space and $\sigma$ are the Pauli matrices. The hopping parameters can be calculated from

$$v_x = \frac{\hbar}{a} t_x \sin(\pm k_0 a), \quad (3)$$

$$v_y = \frac{\hbar}{b} t_y,$$

$$v_z = \frac{\hbar}{c} t_z.$$
and the parameters of $v_x$, $v_y$ and $v_z$ can be calculated from Fermi velocities at Weyl nodes. We calculate the corresponding energy for valence and conduction band in the following form

$$E^{v,c} = \pm \sqrt{A^2 + B^2 + C^2},$$

where $v$ corresponds to the $-$ sign and denotes for the valence band (VB) while $c$ is related to the $+$ sign and indicates to the conduction band (CB). The energy dispersion of TaAs in momentum space as a function of $k_x$ and $k_y$ for $k_z = 0$ is illustrated in Fig. 1(b).

The electron dynamics in an external electric field is coherent since the duration of ultrafast pulse is smaller than the relaxation time of electrons in Weyl semimetals which is a few-picosecond [10]. Then the evolution of the system can be described by time-dependent Schrödinger equation (TDSE)

$$i\hbar \frac{d\psi}{dt} = \mathcal{H}\psi, \quad (5)$$

where the Hamiltonian has explicit time dependence which comes from the time dependent electric field. External electric field causes both interband and intraband electron dynamics. The intraband electron dynamics which describes the redistribution of electron within a band is described by the acceleration theorem [11]

$$\hbar \frac{dk}{dt} = e\mathbf{F}(t). \quad (6)$$

For an electron with initial momentum $\mathbf{q}$, the solution of the Bloch acceleration theorem is

$$\mathbf{k}_F(\mathbf{q}, t) = \mathbf{q} + \frac{e}{\hbar} \int_{-\infty}^{t} \mathbf{F}(t_1) dt_1. \quad (7)$$

This solution does not depend on the band index and therefore the intraband electron dynamics is the same for both conduction and valence band.

The corresponding wave function in the real space are Houston functions [12]

$$\Psi(q, t) = \Psi_{kF}(\mathbf{r}, t) e^{-i\int_{-\infty}^{t} d\tau E_{\alpha}(\mathbf{r}, \tau)}, \quad (8)$$

where $\alpha = v$ for VB and $\alpha = c$ for CB.

The general solution for TDSE in the basis of Houston functions is expressed as

$$\psi_q(r, t) = \sum_{\alpha = v, c} \beta_{\alpha q}(t) \Phi_{\alpha q}(r, t), \quad (9)$$

where $\beta_{\alpha q}(t)$ are expansion coefficients. Let us introduce the following quantities

$$D^{cv}(\mathbf{q}, t) = \mathcal{A}^{cv}[k(\mathbf{q}, t)] \exp \left( i\phi^{(d)}_{cv}(\mathbf{q}, t) \right), \quad (10)$$

$$\phi^{(d)}_{vc}(\mathbf{q}, t) = -\frac{1}{\hbar} \int_{-\infty}^{t} dt_1 (E_c[k(\mathbf{q}, t_1)] - E_v[k(\mathbf{q}, t_1)]), \quad (11)$$

$$\mathcal{A}^{cv}(\mathbf{q}) = \left\langle \Psi_{\mathbf{q}}^{(c)} | i \frac{\partial}{\partial \mathbf{q}} | \Psi_{\mathbf{q}}^{(v)} \right\rangle \quad (12)$$

$$\mathbf{D}^{cv}(\mathbf{q}) = e\mathcal{A}^{cv}(\mathbf{q}) \quad (13)$$

Here $\mathcal{A}^{cv}(\mathbf{q})$ is a matrix element of the non-Abelian Berry connection [13], $\phi^{(d)}_{vc}(\mathbf{q}, t)$ is the dynamic phase, $\Psi_{\mathbf{q}}^{(c)}$ and $\Psi_{\mathbf{q}}^{(v)}$ are the eigenfunctions of the field free Hamiltonian and $\mathbf{D}^{cv}(\mathbf{q})$ is the dipole matrix element which determines the optical transition between the valence band and the conduction band.

The dipole matrix elements can be obtained analytically as

$$D_x = \frac{e}{\sqrt{A^2 + B^2}} \frac{t_x a \sin(k_x a)}{2i(A^2 + B^2 + C^2)} \times [AC + iB \sqrt{A^2 + B^2 + C^2}], \quad (14)$$

$$D_y = \frac{e}{\sqrt{A^2 + B^2}} \frac{t_y b}{2i(A^2 + B^2 + C^2)} \times \left[ C \left( A \sin(k_y b) - B \cos(k_y b) \right) + i\sqrt{A^2 + B^2 + C^2} \right] \times \left( A \cos(k_y b) + B \sin(k_y b) \right), \quad (15)$$

$$D_z = \frac{e}{\sqrt{A^2 + B^2}} \frac{t_z c}{2i(A^2 + B^2 + C^2)} \times \left[ \frac{\sin(k_z c)}{A^2 + B^2} \right] \times (AC + iB \sqrt{A^2 + B^2 + C^2} + \cos(k_z c)). \quad (16)$$

FIG. 1: (Color online) (a) Body-centered tetragonal structure of TaAs. (b) Energy dispersion of TaAs in the reciprocal space obtained with the Hamiltonian [2].

 Houston functions [12] both conduction and valence band.
In these terms, it is more convenient to work in the interaction representation. The Schrödinger equation in the interaction representation can be expressed as

\[ i\hbar \frac{\partial B_q(t)}{\partial t} = H'(q, t)B_q(t), \quad (17) \]

where \( B_q(t) \) and Hamiltonian \( H'(q, t) \) are defined as

\[ B_q(t) = \begin{pmatrix} \beta_{e,q}(t) \\ \beta_{c,q}(t) \end{pmatrix}, \quad (18) \]

\[ H'(q, t) = eF(t)\tilde{A}(q, t), \quad (19) \]

\[ \tilde{A}(q, t) = \begin{bmatrix} 0 & D^{cv}(q, t) \\ D^{cv}(q, t) & 0 \end{bmatrix} \quad (20) \]

A general solution of Schrödinger equation, Eq.\( \text{[17]} \), can be expressed in terms of the evolution operator, \( \tilde{U}(q, t) \), as

\[ B_q(t) = \tilde{U}(q, t)B_q(-\infty), \quad (21) \]

\[ \tilde{U}(q, t) = \tilde{T} \exp \left[ i \int_{t'=-\infty}^{t} \tilde{A}(q, t')dk(q, t') \right], \quad (22) \]

where \( \tilde{T} \) denotes the time-ordering operator and the integral is affected along the Bloch trajectory \( k(q, t) \).

Also, optical pulse induces an electrical current through the system which can be calculated from the following expression

\[ J_j(t) = e \int dq \sum_{a_1=v,c} \sum_{a_2=v,c} \beta_{a_1,q}^* \beta_{a_2,q} V_j^{a_1,a_2} V_j^{a_1,a_2}(q, t), \quad (23) \]

where \( j = x, y, z \) and \( V_j^{a_1,a_2} \) are the matrix elements of the velocity operator, \( V_j = \frac{1}{\hbar} \frac{\partial H}{\partial k} \), between the conduction and valence band states. With the known wave functions for valence and conduction bands the matrix elements of the velocity operator are

\[ V^{y y} = -V^{c c} = \frac{t_y a}{\hbar\sqrt{A^2 + B^2 + C^2}} \times [A \sin(k_y a) - B \cos(k_y a)], \quad (24) \]

\[ V^{y c} = (V^{c y})^* = \frac{t_y g}{\hbar\sqrt{A^2 + B^2 + C^2}} \times \left[ A \sin(k_y a) - B \cos(k_y a) \right] \]

\[ V^{y z} = -V^{c z} = \frac{t_z c}{\hbar\sqrt{A^2 + B^2 + C^2}} \times \left[ A \sin(k_z c) - B \cos(k_z c) \right], \quad (26) \]

\[ V^{z y} = -V^{z c} = \frac{t_y c}{\hbar\sqrt{A^2 + B^2 + C^2}} \times \left[ A \sin(k_z c) + B \cos(k_z c) \right] \]

\[ \times \left[ \cos(k_z c) + \sin(k_z c) \frac{\sin(k_z c)}{A^2 + B^2} \right] \times \left[ AC - iB\sqrt{A^2 + B^2 + C^2} \right]. \quad (27) \]

### III. RESULTS AND DISCUSSION

We use a two-band Bloch Hamiltonian model in three dimensions for TaAs to present topological properties of three-dimensional (3D) Weyl semimetals. TaAs is a body-centered tetragonal lattice system, see Fig.\( \text{[1a]} \), with lattice constants \( a = b = 3.437\text{\AA} \) along \( x \) and \( y \) directions, respectively and \( c = 11.646\text{\AA} \) along \( z \) direction. In this model, see Eq.\( \text{[2]} \), we assume the position of Weyl points \( k_0 = (\pm 0.2, 0, 0) \). Below we apply pulses with different characteristics to investigate the electron dynamics of the system in terms of the electron population of the conduction band in the reciprocal space. Such distribution illustrates topological properties of Weyl semimetals.

#### A. One single oscillation circularly-polarized pulse

We study the response of Weyl semimetals to a single-oscillation circularly polarized pulse in terms of the induction of the CB population in the momentum space. We assume that optical pulse propagate along the \( z \)-direction, see Fig.\( \text{[1a]} \), and its component defines as

\[ F_z(t) = F_0 e^{-u^2} (1 - 2u^2), \quad F_y(t) = 2uF_0 e^{-u^2}, \quad (28) \]

where the field amplitude \( F_0 = 0.01 \text{\text{V/\text{\AA}}} \), \( u = t/\tau \) and \( \tau \) is pulse length which is set, \( \tau = 10 \text{ fs} \). We numerically solve TDSE, see Eq.\( \text{[17]} \) with the initial condition
\((\beta_{q\alpha}, \beta_{\alpha q}) = (1, 0)\). Applied optical field causes transitions of electron from the valence band to the conduction band which resulting a finite CB population. The distribution of the residual conduction band population, \(N_{CB}^{(res)} = |\beta_{\alpha q}(t = \infty)|^2\), the CB population after at the end of the pulse, as a function of \((k_x, k_y, k_z)\) is illustrated in Fig. 3.

We call the Weyl point which is located in the negative direction of \(k_x\), \(k_0 = -0.2 (1/\text{Å})\), \(W\) and the other one in positive direction, \(k_0 = 0.2 (1/\text{Å}), W'\). At \(k_z = 0\), the response of both weyl points to an external electric field are similar and that is exactly the same as what we observed for graphene [15]. Since the interband dipole matrix elements are highly localized near the Weyl points, see Fig. 3, the distribution of electrons has sharp maximum along the separatrix, both outside and inside.

The separatrix, which is shown by a closed black line in Fig. 4 (a) and (b), is a topological object which divides a momentum space into two regions. Any electron Bloch trajectory which originates inside the separatrix encircle the Weyl points and if the Bloch trajectory originates outside the separatrix does not encircle the Weyl points.

The data shows that with an increase of \(k_z\) in positive direction, the \(W\) and \(W'\) become populated differently. For \(W\) point, the major population occurs inside of the separatrix while the minor population occurs outside of the separatrix. In opposite, for \(W'\) the large CB population is located outside the separatrix.

In order to explain the data we turn to Eq. 22. We consider the first order of perturbation theory of evolution operator

\[
\hat{U}(q, t) = 1 + i \int_{t' = -\infty}^t \hat{A}(q, t')dk(q, t').
\]  

(29)

Correspondingly, the residual CB population \(n_\alpha\) is

\[
n_\alpha = \left| \oint |\mathcal{A}_{cv}[k(q, t)]n(t)| \exp \left( i\phi_{cv}^{(tot)}(q, t) \right) dk(q, t) \right|^2,
\]  

(30)

where \(n(t) = F(t)/F(t)\) is the unit vector tangential to the Bloch trajectory and the total phase \(\phi^{(tot)}\) is

\[
\phi_{cv}^{(tot)} = \phi_{cv}^{(T)}(q, t) + \phi_{cv}^{(d)}(q, t),
\]  

(31)

and topological phase is defined as

\[
\phi_{cv}^{(T)}(q, t) = \text{arg} \left( \mathcal{A}_{cv}(k(q, t))n(q, t) \right).
\]  

(32)

Since non-Abelian Berry connection element \(\mathcal{A}_{cv}(k(q, t))\) and topological phase \(\phi_{cv}^{(T)}(q, t)\) are not gauge invariant, they are not observable. However, the CB population \(n_\alpha\) is observable. Fig. 4 (c) and (d) show the topological phase for Weyl nodes with different chirality. For \(q\) outside the separatrix, changes in topological phase is large and closes to \(\pm 2\pi\) while for the point \(q\) inside the separatrix changes in topological phases is much smaller.
$kz < 0$, the topological phase changes its sign and as a result the distribution of electrons at Weyl points is a mirror-symmetric (with respect to the $yz$-plane) of those for $kz > 0$. For left circularly-polarized (LCP) pulse the distribution of the electrons in the conduction band is mirror-symmetric (with respect to $yz$ plane) of that for RCP pulse, and this is due to the chirality selection rule [18].

**B. One single oscillation circularly polarized pulse followed by linear polarized probe pulse**

The texture of the CB population in the momentum space after a circularly-polarized optical pulse, which we explained in the previous section, can be probed by the linear polarized pulse. The probe pulse propagates along the $x$ direction of the system, and its amplitude is $F_0 = 0.002$ (V/Å) which is five times smaller than the amplitude of the main pulse. We apply the probe pulse right after the main circularly polarized pulses at $t = 30$ fs. Fig. 5 (a) and (b) show the electrical current induced by probe pulse in $y$ and $z$ directions. The residual current (the current at $t = 30$ fs) for both RCP and LCP pulses are the same. However, after the linear probe pulse, the residual current (the current at $t = 90$ fs) for both circularly polarized pulse are different and distinguishable.

**C. Two cycles circularly-polarized pulse**

Here we illustrate the chirality of Weyl points in the presence of a circularly-polarized pulse consisting of two cycles with opposite chirality. We use a two-oscillation pulse to present that two Weyl points are chiral in Weyl semimetals. The pulse is incident normally on the system along the $z$ direction and has the following form

\[
F_x(t) = F_0[-e^{-u^2}(1 - 2u^2) + \alpha e^{-(u-u_0)^2}(1 - 2(u - u_0)^2)],
\]

\[
F_y(t) = 2F_0[u e^{-u^2} + \alpha (u - u_0) e^{-(u-u_0)^2}].
\]

Here, $\mp$ determines the circularities (the upper sign is for identical and the lower for opposite circular polarizations). $F_0$ and $\tau$ are the value we explained for a single oscillation pulse. $u_0 = \tau_0/\tau$ and $t_0$ is approximately a half pulse length. We set $t_0 = 60$ fs. We solve the TDSE [17] with initial condition $(\beta_{eq}, \beta_{eq}) = (1, 0)$. Fig. 6 shows the momentum-space interferogram near the Weyl points for a pulse consisting of two optical periods with different circularities. The amplitude, $F_0$, of the second cycle is 0.75%, $\alpha = 0.75$, of the first cycle. The distribution is highly chiral and changing the circularity to the opposite would change the distribution of electron to be mirror reflected in $yz$ plane, see Fig 7. The interferogram at $W$ and $W'$ are different which reflects the intrinsic chirality related to the Berry phase of Weyl semimetals in reciprocal space. At the end of the first cycle of the pulse, $t = 30$ (fs), the conduction band does populate along the separatrix, see Fig. 2, but does not produce any interference fringes. The origin of fringes is related to the passage of the second pulse. During the second optical pulse, whose circularity is opposite of the first optical pulse, clockwise, gradually the formation of interference fringes start in the direction of the electric field rotation along the second separatrix. The origin of chirality is related to the different pathways that an electron moves during the two
FIG. 6: (Color online) Residual CB population as a function of $k$ in reciprocal space for $k_z = 0$, after a two oscillations pulse where the first optical cycle is right-circularly polarized with amplitude $F_0 = 0.01 \, \text{V/Å}$, and the second cycle is left circularly-polarized with amplitude $0.75F_0$.

circular pulse. When the first optical pulse was applied, the electron starts at $k_0$ point in reciprocal space, moves toward the Weyl point, $W$, where the transition between valence and conduction band occurs. At this point there are two possibilities: 1) the electron transfers to the conduction band with the corresponding amplitude $A_1$, 2) the electron stays in the valence band and complete the first cycle in the valence band with amplitude $A_2$ and transfer to the VB during the second pulse. Then for both pathways at the end of the first pulse, the electron returns to the initial points, $k_0$, before continue the second cycle in the opposite direction of the first pulse. If time between the two passages of the $W$ point is minimum, so any dephasing is small and the two amplitudes, $F_0$ and $\alpha F_0$ will interfere and as a result the phase difference between the two amplitudes accumulate the corresponding pathway in reciprocal space. Some part of the fringes are normal to the separatrix which means phase increases fastest along the separatrix. For the $k_0$ points whose the time between the first and second passage of the $W$ point increases up to the optical period, $T$, there is little phase-difference along the separatrices, and the fringes tend to be parallel to the separatrices. This interferometer does not need an external reference source and, therefore, is self-referenced.

Fig. 8 shows the fringes in reciprocal space for a pulse consisting of two subpulses with the same circularity and amplitude. In contrast to Fig. 6, the optical pulse does not cause any interferogram chirality. This is due to the fact that in this case the time between the first and second passage of the electron through the $W$ point is large and is exactly the period $T$, which cause strong dephasing and as a result the fringes is mostly parallel to the separatrix. Also, the field amplitudes for two oscillation are equal and so the probability amplitude of two different possibilities explained above is related as $A_1 = A_2$, causes the distributions to be achiral. However, the distributions for $W_1$ and $W_2$ are still different which is due to the intrinsic chirality of Weyl points in reciprocal space.

FIG. 7: (Color online) Same as Fig.6 but for a pulse with opposite chirality. In the first oscillation electric field rotates clockwise while in the second oscillation does counter-clockwise.

IV. CONCLUSION

In summary, we studied the electron dynamics of Weyl semimetals in a circularly polarized optical pulse with the duration of a few femtoseconds. One single oscillation circularly polarized pulse causes the electrons to circle in the momentum space and accumulate both dynamic phase and topological Berry phase along the separatrix where the maximum transfer between the valence band and conduction band occur. The significant property of the distribution of the electron in momentum space is that for a definite circular pulse, for one Weyl node the inside of the separatrix populates while for the other Weyl node the outside of the separatrix does populate. So in this case, the freedom of an electron to reside at one of the Weyl points, the so-called valley degree of freedom, may also can be considered as a pseudospin, and so they suggest the great potential of Weyl semimetals for applications in spintronics/pseudospintronics. Furthermore, employing a two-cycle circularly polarized pulse
FIG. 8: (Color online) The same as Fig. 6 but for an optical pulse where the two cycles have the same amplitudes and circular polarizations. with opposite chirality and different amplitudes causes the formation of interferogram in reciprocal space which is highly chiral for two Weyl nodes and illustrates the intrinsic chirality of Weyl points in Weyl semimetals.

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