Circular phonon dichroism in Weyl semimetals

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We study the phonon response in magnetic metals with spin-orbit coupling. We show in such systems, phonon Berry curvature and circular phonon dichroism arise. Phonons of left and right circular polarization will be selectively absorbed and have different lifetimes. We investigate the effect in Weyl semimetals. We find that the absorption of left-handed (or right-handed) circularly polarized phonons is much stronger than that of the other one. The effect makes Weyl semimetals potential materials to realize acoustic polarimeters.

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Recent discoveries of topological insulators (TIs) [1, 2] and Weyl semimetals (WSMs) [3–9] have attracted great theoretical and experimental interests because of their nontrivial topological properties and promises for applications such as topological qubits and spintronics. These studies reveal essential roles of spin-orbit coupling (SOC) in realizing novel quantum states of matters. The characterizations of these nontrivial topological states have recently been extended from the electromagnetic response to the viscoelastic or phonon response [10–14]. Actually, non-trivial phonon related phenomenon called the phonon Hall effect (PHE) has already been discovered experimentally [15, 16]. Theoretical analyses relate the PHE to the adiabatic Berry curvature of the electron wave function [17, 18], which is related to the phonon response coefficient in TIs [11–14]. The Berry curvature can be seen as an effective magnetic field for charge neutral phonon, and could have much stronger effects than that for the real magnetic fields.

However, the preceding studies focus on the gapped systems. Similar effects in magnetic metals have not been systematically studied so far. In metals, two problems arise. Firstly, Born-Oppenheimer approximation does not apply since there is no energy gap to preserve electrons from excitation and there is no adiabatic process. Secondly, because in metals phonons could excite electron hole pairs, they will be absorbed while propagating.

In this Letter, we derive the phonon dynamics applicable for magnetic metals. We show that in metals the Berry curvature and the associating effective magnetic field also exist. Furthermore, we find that in such systems, circular phonon dichroism arises. The absorption rates of left-handed circularly polarized (LCP) phonons and right-handed circularly polarized (RCP) phonons are different, meaning the acoustical wave attenuation will depend on the chiralities of the phonons. Similar to the phonon’s effective magnetic field, the circular phonon dichroism is a characteristic phenomenon in magnetic metals with strong SOC.

We apply the theory to Weyl semimetals (WSMs), which contain at least one pair of Weyl nodes of opposite chiralities. We show that a Weyl node will selectively absorb LCP/RCP acoustic phonons, depending on its chirality. In time reversal symmetry (T) broken WSMs, the total effect of a pair of the Weyl nodes remains cancelled. Furthermore the effect can be enhanced by breaking inversion symmetry (I). In such system, one of the two acoustic circularly polarized phonon modes will be attenuated much faster than the other one. Such kind of WSMs can be used to generate circularly polarized phonons.

We start by deriving general formalism for describing effective magnetic field and circular phonon dichroism for metals. In metals, the coupling of electrons and ions is described by the electron-phonon coupling Hamiltonian $H_{ep}$, which is related to the positions of both ions and electrons. We denote the positions of ions as $\{R\} \equiv \{R_n^0 + u_n, n = 1, \ldots, N_i\}$, where $u_n$ is the displacement from the equilibrium position $R_n^0$ and $N_i$ is the total number of ions. For simplicity, we consider a monatomic lattice with atomic mass $M$ and the position of primitive cell can be denoted by $R_n^0$. The resulting formalism can be easily generalized to more general cases.

The problem of lattice vibrations can be solved by the equation of motion:

$$M\ddot{u}_n^\alpha = -\sum_{n',\beta} \Phi_{nn'n'}^\alpha \dot{u}_{n'}^\beta + \langle \hat{T}_{n,\alpha} \rangle$$

where $\alpha, \beta = x, y, z$, and the right hand side of the equation is the total force acting on the $n$-th atom, which contains two parts of contributions: the first term comes from the direct ion-ion interaction with respect to displacement $u_n^\alpha$ under harmonic approximation, and the second term is the force exerted by electrons $\hat{T}_{n,\alpha} = -\partial \hat{H}_{ep}/\partial u_n^\alpha$. $\langle \hat{T}_{n,\alpha} \rangle$ means the expectation value of electron-phonon force in the presence of time-dependent vibrating ion fields. Treating the ions’ movements as the external fields to electrons and $\hat{H}_{ep}(t) \approx \sum_{n\alpha} \left(\partial \hat{H}_{ep}/\partial u_n^\alpha\right) u_n^\alpha(t)$ as perturbation Hamiltonian to the first order of displacements, the electron-ion force can be evaluated using linear response theory.

$$\langle \hat{T}_{n,\alpha}(t) \rangle = -\sum_n \int_{-\infty}^t dt' \mathcal{G}_{n,\alpha}(R_n - R_n', t-t') u_{n'}^\beta(t')$$

(2)
where

\[ G_{\alpha\beta}^{R}(R_n - R_{n'}, t - t') = -\frac{i}{\hbar} \theta(t - t') \times \left\langle \left[ \hat{T}_{n,\alpha}(t), \hat{T}_{n',\beta}(t') \right] \right\rangle_0 \quad (3) \]

In momentum and frequency domain, the equation of motion function is written as

\[ \omega^2 u_q^\alpha = \sum_{\beta} \left[ \Phi_{\alpha\beta}(q) + G_{\alpha\beta}(q, \omega) \right] u_q^\beta \quad (4) \]

\[ \Phi_{\alpha\beta}(q) \] and \( G_{\alpha\beta}(q, \omega) \) are the Fourier transforms of \( \Phi_{n\alpha'}/M \) and \( G_{\alpha\beta}^{R}(R_n, t)/M \), respectively.

We can expand \( G_{\alpha\beta}(q, \omega) \) as a Taylor series in \( \omega \). This is because the characteristic energy of phonons is much smaller than the characteristic energy of electrons, i.e. the Fermi energy \( \varepsilon_F \) : \( \hbar \omega_{CP} \ll \varepsilon_F \). One expects the response function does not change drastically in the energy scale. As a result, \( G_{\alpha\beta}(q, \omega) \) is expanded to:

\[ G_{\alpha\beta}(q, \omega) \approx G_{\alpha\beta}^0(q) + i\omega G_{\alpha\beta}(q) + \omega^2 Z_{\alpha\beta}(q) - 2i\omega \gamma_{\alpha\beta}(q, \omega) \quad (5) \]

The meanings of these terms are explained as follows. The first three terms are the Hermitian part of \( G_{\alpha\beta}(q, \omega) \), expanded in powers of \( \omega \) to the second order. The first term is the electrons’ screening to the ions’ vibration. Together with \( \Phi_{\alpha\beta}(q) \), it gives rise to the dynamical matrix \( D_{\alpha\beta}(q) \), from which one finds the dispersion and eigenmodes of phonons in usual T invariant systems. The second term is the first order of \( G_{\alpha\beta}(q, \omega) \) in \( \omega \), in which the matrix \( G_{\alpha\beta}(q) \) is anti-Hermitian. In gapped insulators, it is easy to show that \( G_{\alpha\beta}(R_n - R_{n'}) \) is exactly the Berry curvature \( 2\hbar i \Im \left\langle \partial \Phi_0 / \partial u_{n'}^\alpha | \partial \Phi_0 / \partial u_n^\beta \right\rangle \big|_{u_n \rightarrow 0} \) defined in Ref.[13]. Therefore, it could be interpreted as the Berry curvature of phonon dynamics, albeit defined for more general systems. The general form of \( G_{\alpha\beta}(q, \omega) \) for non-interacting system is shown in Eq. (4), which could be used to perform the first principles calculation. The third term is a correction to the ion mass. Finally, the last term is the anti-Hermitian part of \( G_{\alpha\beta}(q, \omega) \), and gives rise to the damping (absorption) of phonons.

For non-interacting systems, \( G_{\alpha\beta}(q) \) and \( \gamma_{\alpha\beta}(q, \omega) \) can be expressed as:

\[ G_{\alpha\beta}(q) = \frac{i\hbar}{M} \sum_{ss'} (f_k s - f_{k + q} s') \times \frac{\langle ks | \hat{T}_{q,\alpha} | k + q s' \rangle \langle k + q s' | \hat{T}_{-q,\beta} | ks \rangle}{(\varepsilon_{ks} - \varepsilon_{k+q} s')^2 + \eta^2} \quad (6) \]

\[ \gamma_{\alpha\beta}(q, \omega) = \frac{\pi}{2M\omega} \sum_k (f_k s - f_{k + q} s) \delta (\varepsilon_{ks} - \varepsilon_{k+q} s + \hbar \omega) \times \langle ks | \hat{T}_{q,\alpha} | k + q s \rangle \langle k + q s | \hat{T}_{-q,\beta} | ks \rangle \quad (7) \]

where \( |ks\rangle \) is the single electron state and \( \varepsilon_{ks} \) is the energy dispersion of electrons. \( k \) is the wave vector. \( s, s' \) are the band indexes and in Eq. (7) \( s \) labels the partially-filled band only. \( f_{ks} \) is the Fermi-Dirac distribution. \( \delta(x) \) is the Dirac delta function.

The circular dichroism originates from the non-diagonal part of \( \gamma_{\alpha\beta} \). For three-dimensional media with magnetization field along high symmetry axis (z-axis), \( \gamma_{\alpha\beta} \) has the form

\[ \gamma = \begin{pmatrix} \gamma^D & -i \gamma^A \\ i \gamma^A & \gamma^D \end{pmatrix} \quad (8) \]

In the long wave limit, the left-handed and right-handed circular polarization vectors are \( \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & i & 0 \end{pmatrix}^T \) and \( \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & -i & 0 \end{pmatrix}^T \), respectively. As a result, the linewidths of LCP and RCP phonon modes are \( \gamma^L = \gamma^D + \gamma^A \) and \( \gamma^R = \gamma^D - \gamma^A \). Because the existence of \( \gamma^A \) causes the anisotropy of phonon absorption, we call it the circular dichroism of phonons. The relaxation times of LCP phonons (\( 1/\gamma^L \)) and RCP phonons (\( 1/\gamma^R \)) are different when \( \gamma^A \) is nonzero.

We apply the general formalism to the \( T \) broken WSMs. WSMs are three-dimensional crystals which host pairs of hand touching Weyl nodes in momentum space with opposite chiralities. We investigate the acoustic phonon response in the \( T \) broken WSMs. By solving the problem that Weyl fermions couple to phonons, we can obtain the effective magnetic field and moreover, the damping of phonons.

The electron-phonon coupled Hamiltonian for Weyl fermions could be written as:

\[ H_\pm = \pm \left[ \frac{1}{2} v_F (p_\mu e_\mu^a + e_\mu^a p_\mu) \sigma^a - \lambda \right] + b_3 \sigma^3 \quad (9) \]

with \( a, \mu = 1, 2, 3 \). The subscript \( \pm \) denotes the chirality of the Weyl node with "+" the left-handed chirality and "-" the right handed chirality. \( \sigma^a \) is the the triplet of Pauli matrices. \( e_\mu^a = \delta_\mu^a - \partial_\mu / \partial x_\mu \) is the tetrad (or vielbein) field, which describes the local stretching and rotation and couples the fermion to the deformation \([11, 12]\). The deformation is seen as the change of the underlying geometry here. The corresponding lattice formulation can be found in Ref. [13][14]. The additional term \( b_3 \sigma^3 \) acts as a vector field which breaks the \( T \). It is from the magnetization of the system and not sensitive to the local deformation. It separates the massless Dirac fermion into two Weyl fermions with \( 2k_0 = 2h_3 / \hbar v_F \), and \( 2k_0 \) is the distance that the two Weyl points separate apart in \( z \) direction of the momentum space. \( \lambda \) is a Dresselhaus-like spin-orbit interaction term, which breaks \( T \) but preserves the rotation symmetry around the \( z \) axis [19]. It shifts the two Weyl points to be at different energy levels.

The effective magnetic field for phonons in metallic phase can be calculated with Eq. (6). In the case of WSMs, \( G_{xy}(q) \) is nonzero but divergent because the low energy effective Hamiltonian in Eq. (9) does not give the information of the whole Brillouin zone. Therefore proper renormalization or a...
full lattice calculation is needed. Such problem of divergence that arises from the low energy Hamiltonian has been encountered before \cite{11,12}. Renormalization in Ref. \cite{11} is based on the assumption that the effective magnetic field should depend on the high energy details and it is nonzero even when Chern number is zero. A direct result of nonzero $G_{xy}$ ($q$) is the splitting of the twofold degenerate transverse acoustic (TA) phonon bands. For crystals with high symmetry along the $z$ axis, the previous twofold degenerate TA phonon modes propagating along $z$ direction will be split into two branches, the LCP mode and the RCP mode. The long wavelength behavior can be roughly described by two different acoustic velocities $v_L$ and $v_R$. The schematic dispersions with nonzero $q_z$ are plotted in Fig. 1 with LCP branch represented by dotted red line and RCP branch represented by dotted blue line.

Next, we consider the absorption of phonons in WSMs. A Weyl node is characterized by linear dispersion with a crossing point. The energy bands consist of the upper band and the lower band, which are above and below the Weyl node, respectively. We assume that the Fermi level is in the upper band and the corresponding Fermi wave number is $k_F$. Near the Fermi level a filled electron can absorb a phonon and jump into the empty state. The electron transition can be within the upper band and between the two bands, denoted as inter-band transition and intra-band transition respectively. Because of the laws of energy and momentum conservation, the absorption of the particular phonon modes is forbidden. The allowed momentum-energy regions in which phonons could be absorbed are shown by the shaded area in Fig. 1. Since the acoustic velocity is much smaller than the Fermi velocity, as seen in Fig. 1, only intra-band transition is allowed. It is also right to use Eq. (7), in which only the electron-phonon coupling matrix element within the partially-filled band is reserved.

We first solve the phonon absorption for single Weyl nodes, with $\mathbb{Z}$ preserved ($\lambda = 0$). When the Fermi level is close to the Weyl point, the Fermi wave number $k_F$ is much smaller than $k_0$ and the long wavelength phonons will not induce transitions between different Weyl nodes. Substituting $\partial H_k / \partial u_q^0$ into Eq. (7), the linewidth matrix elements $\gamma^D$ and $\gamma^A$ can be obtained. The leading terms of linewidths for phonon modes with nonzero $q_z$ ($q_x = q_y = 0$) are

$$
\gamma^D_{\pm} = \gamma_0 \left[ \frac{|q_z|}{k_F} + \frac{1}{4} \frac{q_z^2}{k_F^3} \right] + O \left( \frac{\bar{v}_s}{v_F} \right)
$$

and

$$
\gamma^A_{\pm} = \pm \gamma_0 \frac{q_z |q_z|}{k_F^2} + 2 \gamma_0 \frac{q_z^2}{k_F^3} \bar{v}_s \frac{k_F}{k_0} + O \left( \frac{\bar{v}_s^2}{v_F} \right)
$$

where $\gamma_0 = \frac{3}{16} \pi \hbar \left( n_e / \rho_F \right) k_0^2$, and $\bar{v}_s$ is the average TA sound velocity along $z$ direction, $V_F$ the Fermi velocity, $n_e$ the electron number density and $\rho_F$ the ion mass density. The subscript "±" denotes the chirality of Weyl node. Here we have used the relation $n_e = k_0^2 / 3 \pi^2$. $\gamma_0$ shows the strength of phonon damping, and it is proportional to the electron number density. $\gamma^A$ and $\gamma^D$ are both expanded in powers of $\omega_q$, and we neglected the splitting of the TA bands by setting $\omega_{q_z} L/R = \bar{v}_s q_z$.

The anisotropy of phonon absorption of left-handed Weyl node is shown in Fig. 2. Fig. 2 (a) shows the relation of linewidths of LCP and RCP phonons with $q_z$. The relation can be expressed by $\gamma^L/R \approx \frac{1}{2} \gamma_0 \frac{|q_z|}{k_F} \left( q_z / k_F + 2 \right)^2$ approximately, as can be obtained from the leading terms in Eq. (10) and (11). Fig. 2 (b) shows the relative circular dichroism $\gamma^A / \gamma^D$ versus $q_z$. It is an increasing function of $q_z$. The upper limit of wavenumber of phonon mode that can be absorbed is $2k_F$, in which the circular dichroism reaches its maximum of
100% and the absorption is for LCP phonons only. As for the other Weyl node of right-handed chirality "−", from Eq. (11) we can see that the first leading term in $\gamma^A$ will flip sign when the chirality is changed. Therefore the Weyl node with right-handed chirality absorbs RCP phonons better.

Since Weyl nodes always appear in pairs, the total linewidth contributed from a Weyl pair is what can be observed in experiments. Because the right-handed Weyl node can be related to the left-handed Weyl node by $\mathcal{I}$, we have the relation $\gamma^A(q) = \gamma^A(-q)$ (same for $\gamma^D$). The total linewidth should be an even function of $q$, satisfying the inversion symmetry. From Eq. (11), we can find the total circular dichroism $\gamma^A$ of a Weyl pair is $4\gamma_0 (q_z/k_F)^2 (k_F/k_0) (\bar{v}_s/v_F)$. The total $\gamma^D$ is $2\gamma_0 (|q_z|/k_F + \frac{1}{\sqrt{3}} |q_z|/k_F)$. The relation of the resulting linewidths for LCP and RCP phonon modes and relative circular dichroism versus $q_z$ are plotted in Fig. 2 (c-d). Since $k_F$ is much shorter than $k_0$ and $\bar{v}_s$ is much smaller than $v_F$, $\gamma_A$ should be very small compared with $\gamma_D$ and the total circular dichroism is non-vanishing but tiny. The absorption of LCP phonons will be slightly stronger than the RCP phonons. When the vector field reverses direction $b_3 \rightarrow -b_3$, $\gamma_A$ will flip sign. Then the absorption of RCP phonons will be stronger.

The circular phonon dichroism can be greatly enhanced by breaking $\mathcal{I}$. In the case of $\lambda \neq 0$ and zero doping, the two Weyl points of opposite chirality are located at different sides of the Fermi level, $\varepsilon^\pm = \pm \lambda$. In this case, phonons are absorbed by electrons of one Weyl node and holes of the other. Symmetries ensure that the phonon absorption contributed from one node with holes is same with that contributed from the other node with electrons. As a result, the total linewidths contributed from a Weyl pair should be double of that from one single node (Fig. 2 (a)), and the relative circular dichroism $\gamma^A/\gamma^D$ is same with that of a single node (Fig. 2 (b)). So the absorption of LCP phonons is much stronger than the RCP phonons. When $q_z$ approaches to $2k_F$, the absorption rate of RCP phonons approaches to 0, meaning that both of the two Weyl nodes selectively absorb LCP phonons.

Here we consider electron doping. The Fermi level goes up when the electron density increases. As illustrated in the inset of Fig. 3 the chemical potential $\mu$ shows the position of Fermi level. The Fermi wave numbers for "±" Weyl nodes are then $k_F^\pm = 1/\hbar v_F |\lambda \pm \mu|$. For different $\mu$, the relations of total relative circular dichroism $\gamma^A/\gamma^D$ with wave number $q_z$ are plotted in Fig. 3. In the case $0 \leq \mu < \lambda$, the Fermi surface is between the two Weyl nodes, and the relative circular dichroism has a sharp drop at $2k_F^+$ and reaches its maximum at $2k_F^-$. In the case $\mu > \lambda$, the Fermi surface is above the two Weyl nodes. In this case the relative circular dichroism has a sharp rise at $2k_F^-$ and reaches its maximum at $2k_F^-$. The drop and rise are due to the disappearance of the contribution of the "−" Weyl node when $q_z$ reaches $2k_F^-$. When $\mu/\lambda \to 1$ (see $\mu/\lambda = 0.7, 1.3$ in Fig. 3), the Fermi surface reaches the "−" Weyl point. The contribution of the "−" Weyl point is small because of the low density of states, and the phonon absorption is mainly contributed by the "+" Weyl node. Since $\gamma^A$ is always positive, the absorption of LCP phonons is always stronger than RCP phonons. For hole doping with $-\mu$ ($\mu$ is always positive), the result is the same with that of $+\mu$. Therefore with electron or hole doping, the anisotropy of phonons absorption always exists.

A simple way to detect the circular phonon dichroism in ultrasonic experiments is to measure the attenuation of the LCP/RCP acoustical waves. The attenuation lengths for LCP and RCP TA waves will differ by about $(2\bar{v}_s \gamma^A) / (\gamma^D - \gamma^A)$. Another way is to shoot linearly polarized TA waves along the magnetization direction. Due to the effective magnetic field (Eq. (6)), Acoustic Faraday rotation will show up. Moreover, since the phonon absorption rates of LCP and RCP acoustical waves are different, the linearly polarized wave will become elliptically polarized and finally circularly polarized. As a result, $\mathcal{T}$ broken WSMs can be used as acoustic polarimeters to generate circularly polarized acoustic waves. It is worth mentioning that the lattice structure of crystals can also lead to the same effects. The phenomenon is named as acoustical activity (corresponding to the optical activity) [20, 21]. It has been proved some of uniaxial noncentrosymmetric crystal classes are acoustical active, including the Tellurium [21]. Additionally, first-principles calculation proved that trigonal Tellurium under pressure can be regarded as $\mathcal{T}$ broken WSMs [22]. Thus in the experiments that detects the circular dichroism the propagation direction of TA waves should avoid the axes along which acoustic activity shows up.

In summary, we have shown that in magnetic metals with strong SOC, the Berry curvature and the associating effective magnetic field for phonons emerge. Moreover, circular phonon dichroism arises. We proved that this phenomenon exists in $\mathcal{T}$ broken WSMs. We showed that the Weyl node will selectively absorb LCP or RCP phonons. The selective absorption depends on the chirality for a single node and on the magnetization direction for a Weyl pair. Furthermore, this
effect can be enhanced by breaking I. The circular dichroism of phonons makes WSMs very potential materials to realize acoustic polarimeters.

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