


Chiral phonon induced spin polarization

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The current understanding of chirality suggests the existence of a connection between structure and angular momentum, including spin. This is particularly emphasized in the chiral induced spin selectivity effect, where chiral structures act as spin filters. However, the recent discovery of chiral phonons have demonstrated that phonons too may carry angular momentum which also can be regarded as magnetic moments adding to the total moment. Here, it is shown that chiral phonons may induce a nontrivial spin texture in an otherwise nonmagnetic electronic structure. By considering a setup in which electrons and phonons are interfaced with each other, it is shown that chiral phonons may transfer their angular momentum into the electron reservoir which, thereby, becomes spin polarized. It is, moreover, shown that an equivalent mechanism does not exist whenever the electrons are interfaced with achiral phonons.

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Phonons represent the collective nuclear motion within a structure. As such, phonons are the quantum mechanically defined quantities with which the mechanical degrees of freedom are effectively incorporated into the framework of the general quantum field theory. While traditionally being regarded as a quantity carrying linear momentum, it is only recently that angular momentum of phonons have been considered. An incomplete list of important results are phonon Hall, phonon spin Hall, and phonon angular momentum Hall effects [1–7], phonon contribution to spin-relaxation processes [8–11] and the Einstein–de Haas effect [10,12–14], phononically mediated spin-spin interactions [15,16], temperature gradient induced phonon angular momentum [17], and optically activated chiral phonomagnetic effects [18–20]. Experimentally, progress has been made in observations of chiral phonons [21–25] and phonon induced magnetothermal properties [26–28].

The existence of phonon angular momentum opens up the possibility to couple the electronic spin degrees of freedom with the mechanical. It is well established that spin and nuclear motion are coupled directly through, e.g., the Elliot-Yafet mechanism [29–34], but also indirectly via the electronic structure [15], and it has been demonstrated that such coupling opens for a viable explanation of the chiral induced spin selectivity effect [34–37].

Hitherto, however, the angular momentum of phonons and electrons have been considered as separate from one another, where the magnetic moment associated with the chiral phonons has been studied in its own right. While this

is definitely pertinent, angular momentum transfer between phonons and electrons has been discussed in a semiclassical model [38], in which a spin-dependent coupling between phonons and electrons is assumed and it is demonstrated that chiral phonons may give rise to the spin-Seebeck effect, something that was also recently observed in experiments [39]. Nonetheless, the mechanism that enables the angular momentum transfer is yet to be discussed. The purpose of this Letter is to present a coherent theory that ties the existence of phononic angular momentum to a broken electronic spin degeneracy. It is shown that the mechanism is provided through a vibronically assisted spin-orbit interaction and, while this coupling is always present, chiral phonons are required for inducing a spin polarization.

Beginning from the discussion of phonon angular momentum, it can be noticed that chirality is a geometrical property where the structure lacks both inversion and reflection symmetries. It is easy to demonstrate that chiral phonons must carry a nonvanishing angular momentum \mathbf{J}_{ph} . This can be seen directly from the definition

$$\mathbf{J}_{\text{ph}}(t) = \int \mathbf{Q}(\mathbf{r}, t) \times \dot{\mathbf{Q}}(\mathbf{r}, t) d\mathbf{r}, \quad (1)$$

where \mathbf{Q} is the nuclear displacement, which is connected to phonons through the relation $\mathbf{Q}(\mathbf{r}, t) = \sum_q l_q \epsilon_q Q_p(t) e^{i\mathbf{q}\cdot\mathbf{r}}$, where $Q_q = b_{\mathbf{q}\mu} + b_{\mathbf{q}\mu}^\dagger$ is the quantum phonon displacement operator. Here, b_q and b_q^\dagger denote the phonon destruction and creation operators, respectively, with $q = (\mathbf{q}, \mu)$ [$\bar{q} = (\bar{\mathbf{q}}, \mu) = (-\mathbf{q}, \mu)$] comprising the wave vector \mathbf{q} and normal mode μ , and $l_q = \sqrt{\hbar/2\rho v \omega_q}$, which defines a length scale in terms of the phonon energy ω_q , density ρ , and volume v , whereas ϵ_q is the displacement polarization vector.

Hence the expectation value of the angular momentum assumes the form

$$\langle \mathbf{J}_{\text{ph}} \rangle(t) = \lim_{t' \rightarrow t} i\partial_{t'} \sum_{pq} l_p l_q \epsilon_p \times \epsilon_{\bar{q}} D_{pq}^>(t', t) \int e^{i(\mathbf{p}-\mathbf{q})\cdot\mathbf{r}} d\mathbf{r}, \quad (2)$$

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where $D_{pq}^>(t', t) = (-i)\langle Q_p(t')Q_{\bar{q}}(t) \rangle$ defines the correlations between the phonons $Q_p(t')$ and $Q_{\bar{p}}(t)$. The expression in Eq. (2) shows that a finite phonon angular momentum requires noncollinear polarizations ϵ_p and $\epsilon_{\bar{q}}$.

Noncollinear polarizations can be achieved when there is a mechanism that mixes the phonon modes p and q . Such mode mixing may originate from, e.g., anharmonic effects or scattering off defects acting upon the otherwise orthogonal modes defined in the harmonic approximation.

In the harmonic approximation, the phonons can, up to a constant, be summarized in a Hamiltonian form as $\mathcal{H}_{\text{ph}} = \sum_q \omega_q b_q^\dagger b_q$. In this form, it is assumed that the phonon modes do not mix which, therefore, leads to a vanishing angular momentum. However, the introduction of a component of the kind $W_{\mathbf{q}\mu\nu} b_{\mathbf{q}\mu}^\dagger b_{\mathbf{q}\nu}$ provides a mode mixing, which can be understood as chirality.

To see this, consider the phonon spinor $\Phi_{\mathbf{q}} = \{b_{\mathbf{q}\mu_i}\}_{i=1}^N$, for N modes, which enables us to write the phonon model as

$$\mathcal{H}_{\text{ph}} = \sum_{\mathbf{q}} \Phi_{\mathbf{q}}^\dagger \omega_{\mathbf{q}} \Phi_{\mathbf{q}}. \quad (3)$$

Written like this, the phonon spectrum is defined through the matrix $\omega_{\mathbf{q}} = \omega_{0\mathbf{q}}\tau^0 + \omega_{1\mathbf{q}} \cdot \boldsymbol{\tau}$, where $\omega_{0\mathbf{q}}$ and $\omega_{1\mathbf{q}}$ represent the mode conservative and mode mixing components, respectively, whereas τ^0 and $\boldsymbol{\tau}$ are the N -dimensional identity and vector of spin matrices. While in this model it is assumed that the mixing only takes place between modes μ_i and μ_j with the same momentum \mathbf{q} , it is straightforward to generalize the model to also include mixing between different momenta.

Considering a structure with two modes, such that $\boldsymbol{\tau}$ reduces to the Pauli matrices, the phonon Green's function $\mathbf{D}_{\mathbf{q}}(z) = \langle\langle \Phi_{\mathbf{q}} | \Phi_{\bar{\mathbf{q}}} \rangle\rangle(z)$ can be written as

$$\mathbf{D}_{\mathbf{p}}(z) = 2\omega_{\mathbf{q}} \frac{z^2 - \omega_{0\mathbf{q}}^2 - \omega_{1\mathbf{q}}^2 + 2\omega_{0\mathbf{q}}\omega_{1\mathbf{q}} \cdot \boldsymbol{\tau}}{(z^2 - \omega_{0\mathbf{q}}^2 - \omega_{1\mathbf{q}}^2)^2 - 4\omega_{0\mathbf{q}}^2\omega_{1\mathbf{q}}^2} \quad (4a)$$

$$= \frac{1}{2} \sum_{s=\pm 1} \frac{2\omega_{\mathbf{q}s}}{z^2 - \omega_{\mathbf{q}s}^2} (\tau^0 + s\hat{\omega}_{1\mathbf{q}} \cdot \boldsymbol{\tau}), \quad (4b)$$

where $\omega_{\mathbf{q}s} = \omega_{0\mathbf{q}} + s\omega_{1\mathbf{q}}$, $\omega_{1\mathbf{q}} = |\omega_{1\mathbf{q}}|$, and $\hat{\omega}_{1\mathbf{q}} = \omega_{1\mathbf{q}}/\omega_{1\mathbf{q}}$. The form of this propagator written in Eq. (4) explicitly describes two modes with opposite helicity or chirality.

By contrast, the polarization for a chiral mode is neither reflection nor inversion symmetric, where the latter condition leads to that $\epsilon_{\mathbf{q}\mu}^* = \epsilon_{\bar{\mathbf{q}}\mu} \neq \epsilon_{\mathbf{q}\mu}$, since equality in the last relation requires inversion symmetry. For, e.g., a helical structure with transversal and longitudinal lattice parameters a and c , respectively, the polarization may be expressed as $\epsilon_{\mathbf{q}} = (a \cos \phi_q, a \sin \phi_q, \phi_q)/d(\phi_q)$, where $\phi = c/2\pi$ and $d(\phi_q) = \sqrt{a^2 + \phi^2 \phi_q^2}$, which displays a variation of the mode that depends on the azimuthal angle ϕ_q . Hence, for such a mode, the vector product $\epsilon_{\mathbf{q}} \times \epsilon_{\bar{\mathbf{q}}} = (-a \phi \phi_q \sin \phi_q, a \phi \phi_q \cos \phi_q, -a^2 \sin 2\phi_q)/d^2(\phi_q)$, which suggests that a free chiral phonon mode, for which $D_{\mathbf{q}}^>(t, t') = (-i)[n_B(\omega_{\mathbf{q}})e^{-i\omega_{\mathbf{q}}(t-t')} - n_B(-\omega_{\mathbf{q}})e^{i\omega_{\mathbf{q}}(t-t')}]$, where $n_B(\omega)$ is the Bose-Einstein distribution function, carries the

nonvanishing angular momentum

$$\langle \mathbf{J}_{\text{ph}} \rangle = \frac{\omega_{\mathbf{q}} l_{\mathbf{q}}^2}{\pi d^2(\phi_q)} \begin{pmatrix} -a \phi \phi_q \sin \phi_q \\ a \phi \phi_q \cos \phi_q \\ -a^2 \pi \sin 2\phi_q \end{pmatrix}. \quad (5)$$

It is important to notice that the off-diagonal components of the phonon propagator, Eq. (4), carry the phase factors $e^{\pm i\phi_p}$, where ϕ_p defines the azimuthal angle of the momentum vector \mathbf{p} . This phase dependence reoccurs also in the helical polarization vector and is an essential feature of chirality. Due to this phase dependence, the phonons necessarily carry angular momentum.

As the above discussion is made plausible, angular momentum can quite naturally be associated with chiral phonons. The objective in this Letter is to show that such phononic angular momentum can be transferred to the electronic subsystem and, hence, induce a spin polarization. For this sake, consider the coupling between electrons and phonons, which generally can be written as [34]

$$\mathcal{H}_{\text{e-ph}} = \sum_{\mathbf{k}\mathbf{q}} \psi_{\mathbf{p}+\mathbf{k}}^\dagger \mathbf{U}_{\mathbf{k}\mathbf{q}} \psi_{\mathbf{k}} Q_{\mathbf{q}}, \quad (6)$$

where the coupling matrix $\mathbf{U}_{\mathbf{k}\mathbf{q}} = U_{\mathbf{k}\mathbf{q}}\sigma^0 + \mathbf{J}_{\mathbf{k}\mathbf{q}} \cdot \boldsymbol{\sigma}$ accounts for a spin-conservative electron-phonon coupling, $U_{\mathbf{k}\mathbf{q}}$, and an electron-phonon assisted spin-orbit interaction, $\mathbf{J}_{\mathbf{k}\mathbf{q}}$.

The spin polarization $\langle \mathbf{M}_{\mathbf{k}} \rangle$ of the electrons are calculated using the identity $\langle \mathbf{M}_{\mathbf{k}} \rangle = (-i)\text{sp}\sigma \int \mathbf{G}_{\mathbf{k}\mathbf{k}'}^<(\omega)d\omega/4\pi$, where $\mathbf{G}_{\mathbf{k}\mathbf{k}'}^<(\omega)$ defines the lesser form of the general single electron Green's function $\mathbf{G}_{\mathbf{k}\mathbf{k}'}(z) = \langle\langle \psi_{\mathbf{k}} | \psi_{\mathbf{k}'}^\dagger \rangle\rangle(z)$, whereas sp denotes the trace over spin-1/2 space. To the second order (Hartree-Fock) approximation in the electron-phonon coupling, this Green's function can be calculated from the Dyson equation

$$\mathbf{G}_{\mathbf{k}\mathbf{k}'}(z) = \delta(\mathbf{k} - \mathbf{k}')\mathbf{g}_{\mathbf{k}}(z) + \mathbf{g}_{\mathbf{k}}(z) \sum_{\mathbf{k}''} \Sigma_{\mathbf{k}\mathbf{k}''}^{(\text{HF})}(z)\mathbf{G}_{\mathbf{k}''\mathbf{k}'}(z), \quad (7a)$$

$$\Sigma_{\mathbf{k}\mathbf{k}'}^{(\text{HF})}(z) = (-i)\delta(\mathbf{k} - \mathbf{k}') \sum_{\mathbf{q}\mathbf{q}'} D_{\mathbf{q}\mathbf{q}'} \mathbf{U}_{\mathbf{k}\mathbf{q}} \text{sp} \int \mathbf{G}_{\mathbf{k}\mathbf{k}'}^<(\omega) \mathbf{U}_{\mathbf{k}\mathbf{q}'} \frac{d\omega}{2\pi} - \frac{1}{\beta} \sum_{z\nu\mathbf{q}\mathbf{q}'} \mathbf{U}_{\mathbf{k}\mathbf{q}} \mathbf{G}_{\mathbf{k}\mathbf{k}'}(z+z_\nu) \mathbf{U}_{\mathbf{k}\mathbf{q}'} D_{\mathbf{q}\mathbf{q}'}(z_\nu), \quad (7b)$$

where $D_{\mathbf{q}\mathbf{q}'} = \int \delta(\omega) D_{\mathbf{q}\mathbf{q}'}^a(\omega)d\omega$.

While this equation should be self-consistently solved, for an analysis of the induced spin symmetries, it is sufficient to replace the electronic Green's functions in these expressions with its unperturbed form $\mathbf{g}_{\mathbf{k}}(z) = \sigma^0 \mathbf{g}_{\mathbf{k}}(z)$, where $\mathbf{g}_{\mathbf{k}}(z) = 1/(z - \epsilon_{\mathbf{k}})$ and $\mathbf{g}_{\mathbf{k}}^<(\omega) = 2\pi\sigma^0 f(\omega)\delta(\omega - \epsilon_{\mathbf{k}})$, whereas $f(\omega)$ is the Fermi-Dirac distribution function. These replacements lead to the simplified self-energy

$$\Sigma_{\mathbf{k}}^{(\text{HF})}(z) = 2f(\epsilon_{\mathbf{k}}) \sum_{\mathbf{q}\mathbf{q}'} \delta(\mathbf{q}) U_{\mathbf{k}\mathbf{q}} D_{\mathbf{q}\mathbf{q}'} \mathbf{U}_{\mathbf{k}\mathbf{q}'} - \frac{1}{\beta} \sum_{\nu\mathbf{q}\mathbf{q}'} \mathbf{g}_{\mathbf{k}-\mathbf{q}}(z - z_\nu) \mathbf{U}_{\mathbf{k}\mathbf{q}} D_{\mathbf{q}\mathbf{q}'}(z_\nu) \mathbf{U}_{\mathbf{k}\mathbf{q}'} \quad (8)$$

The Hartree (first) contribution is to lowest order linear in the component $\mathbf{J}_{\mathbf{k}\mathbf{q}} \cdot \boldsymbol{\sigma}$ which, hence, shows that the coupling

between the electrons and phonons may break time-reversal symmetry. However, this contribution vanishes in this approximation since there are no phonons at $\mathbf{p} = 0$. By contrast, the exchange (second) contribution does not only open for breaking the time-reversal symmetry, since in general

$$\mathbf{U}_{kp}\mathbf{U}_{kq} = U_{kq}U_{kq'} + \mathbf{J}_{kq} \cdot \mathbf{J}_{kq'} + (U_{kq}\mathbf{J}_{kq'} + \mathbf{J}_{kq}U_{kq'} + i\mathbf{J}_{kq} \times \mathbf{J}_{kq'}) \cdot \boldsymbol{\sigma}, \quad (9)$$

but it also provides a correlation between the electronic and phononic degrees of freedom.

However, before showing that chiral phonon leads to a breaking of the electronic spin degeneracy, it is pertinent to discuss the origin of the phonon induced spin-polarization and spin-flip processes which are enabled by \mathbf{J}_{kq} . For this purpose, it is instructive to consider the spin-orbit coupling, which can be written as [40]

$$H_{\text{SOC}} = \frac{\xi}{2} [\mathbf{E} \times \mathbf{p} - \mathbf{p} \times \mathbf{E}] \cdot \boldsymbol{\sigma}, \quad (10)$$

where \mathbf{E} is the total electric field acting on the electrons, whereas $\xi = 1/4c^2$ in atomic units. By introducing the field operator $\psi(\mathbf{r}) = \int \psi_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} d\mathbf{k}/\Omega$ and its Hermitian conjugate, and assuming time-independent magnetic fields \mathbf{B} , such that $\nabla \times \mathbf{E} = -\partial_t \mathbf{B} = 0$, the spin-orbit coupling contribution can in the second quantization be written

$$\mathcal{H}_{\text{SOC}} = \mathcal{E}_S - i\xi \int \psi_{\mathbf{k}}^\dagger(\mathbf{k} \times \mathbf{k}') \cdot \boldsymbol{\sigma} V(\mathbf{r}) e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}} \psi_{\mathbf{k}'} d\mathbf{r} \frac{d\mathbf{k}}{\Omega} \frac{d\mathbf{k}'}{\Omega}, \quad (11)$$

where \mathcal{E}_S is the boundary surface contribution and where the electric field has been identified with the electronic confinement potential $V(\mathbf{r})$ using the relation $\mathbf{E} = -\nabla V(\mathbf{r})$.

Here, $\mathcal{E}_S = \int \psi_{\mathbf{k}} \mathcal{S}_{\mathbf{k}\mathbf{k}'} \psi_{\mathbf{k}'} d\mathbf{k} d\mathbf{k}'/\Omega^2$, where $\mathcal{S}_{\mathbf{k}\mathbf{k}'}$ is given by

$$\mathcal{S}_{\mathbf{k}\mathbf{k}'} = \xi \int_S (\mathbf{k} \times \hat{\mathbf{n}}) \cdot \boldsymbol{\sigma} V(\mathbf{r}) e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}} dS, \quad (12)$$

where $\hat{\mathbf{n}}$ denotes the outward normal on the boundary surface \mathcal{S} . This contribution can be identified as the Rashba spin-orbit coupling.

The confinement potential $V(\mathbf{r}) = \int V(\mathbf{r} - \mathbf{r}') d\mathbf{r}'$, where $\mathbf{r} = \mathbf{r}_0 + \mathbf{Q}(\mathbf{r})$ denotes the coordinate for the nuclei in terms of the equilibrium position \mathbf{r}_0 and displacement $\mathbf{Q}(\mathbf{r})$. For small displacements, the potential can be expanded around the equilibrium positions $V(\mathbf{r} - \mathbf{r}') \approx V(\mathbf{r} - \mathbf{r}_0) - \mathbf{Q} \cdot [\nabla_{\mathbf{r}'} V(\mathbf{r} - \mathbf{r}')]_{\mathbf{r}' \rightarrow \mathbf{r}_0}$, where the first contribution along with the surface integral provide the static spin-orbit interaction.

The spin-orbit interaction with the lowest order coupling to the nuclear displacement is given by the Hamiltonian

$$\mathcal{H}_{\text{SOC}}^{(1)} = (-i)\hbar\xi \int \psi_{\mathbf{k}}^\dagger [(\mathbf{k} \times \mathbf{k}') \cdot \boldsymbol{\sigma}] [\mathbf{Q} \cdot [\nabla_{\mathbf{r}'} V(\mathbf{r} - \mathbf{r}')]_{\mathbf{r}' \rightarrow \mathbf{r}_0}] \times e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}} \psi_{\mathbf{k}'} d\mathbf{r}_0 d\mathbf{r} \frac{d\mathbf{k}}{\Omega} \frac{d\mathbf{k}'}{\Omega}. \quad (13)$$

Using $\nabla_{\mathbf{r}'} V(\mathbf{r} - \mathbf{r}') = (-i) \int \mathbf{q} V(\mathbf{q}) e^{i\mathbf{q}\cdot(\mathbf{r}-\mathbf{r}')} d\mathbf{q}/\Omega$ leads to the phonon assisted spin-orbit interaction being

written as

$$\mathcal{H}_{\text{SOC}}^{(1)} = \int \psi_{\mathbf{k}+\mathbf{q}}^\dagger \mathbf{J}_{\mathbf{k}\mathbf{q}} \cdot \boldsymbol{\sigma} Q_{\mathbf{q}} \psi_{\mathbf{k}} \frac{d\mathbf{q}}{\Omega} \frac{d\mathbf{k}}{\Omega}, \quad (14)$$

with $\mathbf{J}_{\mathbf{k}\mathbf{q}} = i\xi U_{kq}(\mathbf{k} \times \mathbf{q})$, where $U_{kq} = U_q = i l_q \epsilon_q \cdot \mathbf{q} V(\mathbf{q})$ is the spin-conservative electron-phonon interaction. This gives the total electron-phonon coupling $\mathbf{U}_{kq} = U_q \sigma^0 + \mathbf{J}_{kq} \cdot \boldsymbol{\sigma}$.

From an order of magnitude estimation, it can be deduced that the ratio of the exchange diagrams originating from the spin-dependent and spin-conservative processes, that is, those which are proportional to $U_{kq}\mathbf{J}_{kq}$ and $U_{kq}U_{kq}$, respectively, is roughly $2\xi q_c/3 (\times \mathbf{k})$, where q_c is the momentum corresponding to the phononic high energy cutoff.

More importantly, however, is that while \mathbf{J}_{kq} is linear in the spin-orbit coupling parameter ξ , that is, in the same order as the static spin-orbit interaction contribution $\mathcal{H}_{\text{SOC}}^{(0)}$, the phonon assisted contribution may potentially have a deeply profound influence on the electronic structure. The reason is that the coupling to phonons by all means generates many-body effects, that is, electronic correlations, which may lead to spontaneous symmetry breaking of the electrons.

To see this, consider the influence from the chiral phonons on the magnetic moment $\langle \mathbf{M} \rangle = \langle \mathbf{M}_{\mathbf{k}} \rangle d\mathbf{k}/\Omega$. In the first order approximation of the lesser Green's function, $\mathbf{G}_{\mathbf{k}}^< \approx \mathbf{g}_{\mathbf{k}}^< + \mathbf{g}_{\mathbf{k}}^r \Sigma_{\mathbf{k}}^{\text{(HF),<}} \mathbf{g}_{\mathbf{k}}^a$, the \mathbf{k} -resolved spin moment reduces to the expression $[\Omega = (2\pi)^3]$

$$\langle \mathbf{M}_{\mathbf{k}} \rangle = 4\xi \int f(\epsilon_{\mathbf{k}}) I_q^2 V^2(\mathbf{q}) \mathbf{K}_{\mathbf{k}\mathbf{q}} \frac{d\mathbf{q}}{(2\pi)^3}, \quad (15a)$$

$$\mathbf{K}_{\mathbf{k}\mathbf{q}} = |\epsilon_q \cdot \mathbf{q}|^2 (\mathbf{k} \times \mathbf{q}) \sum_{ss'=\pm 1} s' n_B(s' \omega_{\mathbf{q}s}) \times \frac{1 + s \hat{\omega}_{\perp\mathbf{q}} \cos \phi_q}{(\epsilon_q + s' \omega_{\mathbf{q}s} - \mathbf{k} \cdot \mathbf{q})^2}, \quad (15b)$$

where $\hat{\omega}_{\perp\mathbf{q}} = |(\omega_{1\mathbf{q}}^{(x)}, \omega_{1\mathbf{q}}^{(y)}, 0)|/\omega_{1\mathbf{q}}$.

By integrating out the phases ϕ_q and θ_q , defining the spin texture $\tilde{\mathbf{K}}_{\mathbf{k}\mathbf{q}} = \int_0^\pi \int_0^{2\pi} \mathbf{K}_{\mathbf{k}\mathbf{q}} d\phi_q \sin \theta_q d\theta_q / (2\pi)^2$, under the assumption that $V(\mathbf{q}) = V(|\mathbf{q}|)$, the resulting kernel is a regular function of the electronic momentum \mathbf{k} . This is shown in Fig. 1, where $\tilde{\mathbf{K}}_{\mathbf{k}\mathbf{q}}$ is plotted as a function of the polar and azimuthal angles θ_k and ϕ_k , respectively, for (a)–(c) achiral and (d)–(f) chiral phonons.

Both for achiral and chiral phonons, there is a vibrationally assisted spin-orbit coupling induced in the electronic structure, which follows on general grounds and has been discussed in different contexts, e.g., Ref. [41]. The absence of a z component for achiral phonons in the example given in Fig. 1 can be understood through the following observation.

The denominator in $\mathbf{K}_{\mathbf{k}\mathbf{q}}$, see Eq. (15b), depends on the difference between the phononic and electronic phases ϕ_q and ϕ_k , respectively, as $[A_{\mathbf{k}\mathbf{q}ss'} - B_{\mathbf{k}\mathbf{q}} \cos(\phi_q - \phi_k)]^2$, where $A_{\mathbf{k}\mathbf{q}ss'} = q^2/2 + s' \omega_{\mathbf{q}s} - kp \cos \theta_k \cos \theta_q$ and $B_{\mathbf{k}\mathbf{q}} = kp \sin \theta_k \sin \theta_q$. This is an even function of the phase difference $\phi_q - \phi_k$ on the interval $[0, 2\pi]$, while both $|\epsilon_q \cdot \mathbf{q}|^2$ and $\mathbf{k} \times \mathbf{q}$ can be written as a sum of even and odd functions of $\phi_q - \phi_k$. For the polarization $\epsilon_q = (0, 0, 1)$, the numerator of the longitudinal spin texture $K_{\mathbf{k}\mathbf{q}}^{(z)}$ is proportional to $\sin(\phi_q - \phi_k)$, which is odd on the interval $[0, 2\pi]$. Hence $\int_0^{2\pi} K_{\mathbf{k}\mathbf{q}}^{(z)} d\phi_q / 2\pi = 0$. For

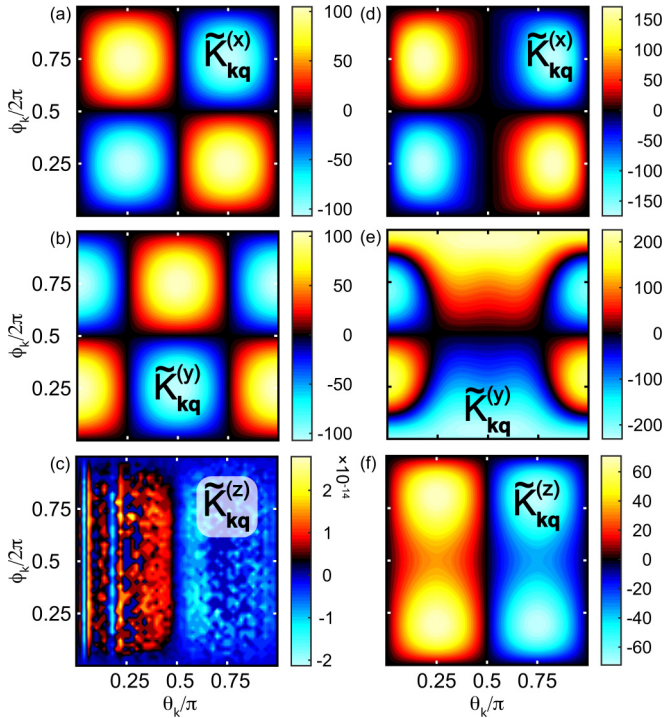


FIG. 1. Examples of $\tilde{\mathbf{K}}_{\mathbf{k}\mathbf{q}} = (\tilde{K}_{\mathbf{k}\mathbf{q}}^{(x)}, \tilde{K}_{\mathbf{k}\mathbf{q}}^{(y)}, \tilde{K}_{\mathbf{k}\mathbf{q}}^{(z)})$ as functions of the polar (horizontal) and azimuthal (vertical) angles θ_k and ϕ_k , for (a)–(c) achiral and (d)–(f) chiral phonons. Parameters used are (a.u.) $k = \sqrt{2}m_e\varepsilon/\hbar$, $\varepsilon = 0.15$ eV, $\omega_{\mathbf{q}\mathbf{s}} = (\omega_0 + s\omega_1)\mathbf{q}$, $\omega_0\mathbf{q} = 0.04$ eV, $\omega_0 = 1$, $\omega_1 = \omega_0/10$, and $\boldsymbol{\epsilon} = (0, 0, 1)$ at $T = 300$ K.

chiral phonons, $\hat{\omega}_{\perp\mathbf{q}} \neq 0$, which leads to the numerator of $K_{\mathbf{k}\mathbf{q}}^{(z)}$ also containing a term which is proportional to $\sin(\phi_q - \phi_k) \cos \phi_q = \sin(\phi_q - \phi_k)[\cos \phi_k \sin(\phi_q - \phi_k) + \sin \phi_k \cos(\phi_q - \phi_k)]$, where the first (second) contribution is an even (odd) function of $\phi_q - \phi_k$. Therefore, $\int_0^{2\pi} K_{\mathbf{k}\mathbf{q}}^{(z)} d\phi_q/2\pi \neq 0$ for chiral phonons.

The above discussion pertains to longitudinally polarized phonons. For transversely (xy -plane) polarized phonons, the vibrationally assisted spin-orbit coupling generates a non-vanishing spin texture for all $\tilde{K}_{\mathbf{k}\mathbf{q}}^{(i)}$, $i = x, y, z$. However, the longitudinal spin texture $\tilde{K}_{\mathbf{k}\mathbf{q}}^{(z)}$ induced for achiral phonons polarized along some transverse axis, e.g., $\hat{\mathbf{x}}$, exactly cancels the spin texture induced for phonons polarized along its orthogonal transverse axis, e.g., $\hat{\mathbf{y}}$. Chiral phonons, on the other hand, break this symmetry such that there remains a net longitudinal spin texture after summing over all phonon modes.

The conclusion is that a vibrationally assisted spin-orbit coupling exists for any type of phonon which, in turn, gives rise to a transversal spin texture. However, a net longitudinal spin texture only arises in the presence of chiral phonons. Nevertheless, as is illustrated in Fig. 1, no magnetic moment is generated from this spin texture since its angular dependencies integrate to zero.

A manifestation of the chiral phonon induced spin texture is the presence of spin currents. The spin current can, in the current approximation, be written as $\mathbb{J}(\mathbf{r}) = (-i)\text{sp}\sigma \int \mathbf{k}\mathbf{G}_{\mathbf{k}}^<(\omega)d\mathbf{k}d\omega/4\pi\Omega = \int \mathbf{k}\langle\mathbf{M}_{\mathbf{k}}\rangle d\mathbf{k}/\Omega$, where the last expression vividly suggests a drift of the spin texture. This drift is a consequence of the spin-orbit coupling and exists

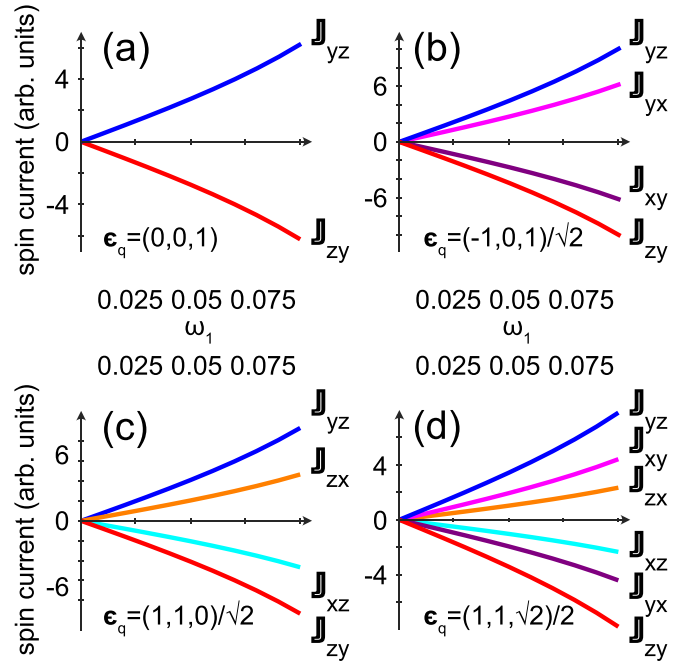


FIG. 2. Nonzero components of the spin-current tensor as functions of the mixing ω_1 for phonons polarized along (a) $\boldsymbol{\epsilon}_{\mathbf{q}} = (0, 0, 1)$, (b) $\boldsymbol{\epsilon}_{\mathbf{q}} = (-1, 0, 1)/\sqrt{2}$, (c) $\boldsymbol{\epsilon}_{\mathbf{q}} = (1, 1, 0)/\sqrt{2}$, and (d) $\boldsymbol{\epsilon}_{\mathbf{q}} = (1, 1, \sqrt{2})/2$. Other parameters are as in Fig. 1.

under general conditions, including equilibrium. However, a net spin flow can only be induced by chiral phonons, not by achiral phonons. This is illustrated in Fig. 2, where the nonzero components of the spin-current tensor are plotted as functions of the mode mixing parameter ω_1 for different phonon polarizations. Achiral phonons are represented by $\omega_1 = 0$, at which condition there is no induced spin current, regardless of the phonon polarization.

By contrast, chiral phonons induce spin currents for which the spatial distribution depends on the polarization. As can be seen in Fig. 2, for phonons polarized solely in the longitudinal (or a transverse) direction, the spin currents flow between this and one other direction only. For phonons polarized along some direction with both longitudinal and transverse components, the spin currents flow between all these. However, in general, the diagonal spin currents, \mathbb{J}_{ii} , $i = x, y, z$, are negligible, which is expected for a noncollinear spin texture.

It should be noticed that the spin currents induce charge currents J_i , owing to the relation $J_i \propto \epsilon_{ijk}\mathbb{J}_{jk}$ [42], where ϵ_{ijk} is the Levi-Civita symbol. Therefore, the spin currents in, e.g., Fig. 2(b) induce a charge current $\mathbf{J} = (J_x, 0, J_z)$, where $J_x \propto \mathbb{J}_{yz} - \mathbb{J}_{zy}$ and $J_z \propto \mathbb{J}_{xy} - \mathbb{J}_{yx}$. There may, hence, be a spin-polarized drift of the charge in the xz plane of the structure.

The induced spin currents should be measurable as a spin accumulation at, e.g., a terminating surface, by exposing the system to an accelerating force. For instance, a spatially weakly inhomogeneous temperature, such that $T = T(\mathbf{r}) \approx T_0 + (\mathbf{r} - \mathbf{r}_0) \cdot \nabla_{\mathbf{r}}T(\mathbf{r})_{\mathbf{r}_0}$, where $T_0 = T(\mathbf{r}_0)$ represents the reference temperature, provides an accelerating field acting on the electrons via the temperature gradient ∇T . Then, to linear order in this field, the longitudinal

spin-current partitions into two contributions. The first of these derive from the electronic temperature variations $k_B\beta^2([\varepsilon_{\mathbf{k}} - \varepsilon_F]/4)\cosh^{-2}(\beta[\varepsilon_{\mathbf{k}} - \varepsilon_F]/2)(\mathbf{r} - \mathbf{r}_0) \cdot \nabla T$. This contribution gives the expected drift of electrons around the Fermi level ε_F .

The second contribution derives from the thermal variations in the phonon distribution, providing accelerating forces involving the processes $\sum_{ss'} k_B\beta^2\omega_{\mathbf{q}s}(1 + s\omega_{\perp\mathbf{q}}\cos\phi_{\mathbf{q}})[2\sinh(s'\beta\omega_{\mathbf{q}s}/2)(\varepsilon_{\mathbf{q}} + s'\omega_{\mathbf{q}s} - \mathbf{k} \cdot \mathbf{q})]^{-2}(\mathbf{r} - \mathbf{r}_0) \cdot \nabla T$. The factor s' makes reference to the absorption ($s' = -1$) and emission ($s' = 1$) processes, whereas s refers to the effective chirality. Ultimately, the phonon driven contribution to the drift current results from the net of the two opposite chiralities; cf. Eq. (4). The theoretical model predicts that the induced spin currents should result in a spin accumulation at some interface surface intersecting the current, in agreement with recent experimental observations [39], where a spin accumulation was measured using the magneto-optical Kerr effect.

In conclusion, it has been demonstrated that the angular momentum carried by chiral phonons may be transferred to electrons via a phase exchange. The phonon chirality translates to the electron spin in the sense that it generates a nontrivial spin texture including circulating spin currents. First principles calculations of interfaces between real chiral insulators and metals should shed additional light on the nature of the discussed phenomenology. While, thus far, out of reach within the realms of state-of-the-art computational methods, the present study will hopefully justify such developments.

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