Chiral phonons and pseudoangular momentum in nonsymmorphic systems

Tiantian Zhang and Shuichi Murakami *Department of Physics, Tokyo Institute of Technology, Ookayama, Meguro-ku, Tokyo 152-8551, Japan
and Tokodai Institute for Element Strategy, Tokyo Institute of Technology, Nagatsuta, Midori-ku, Yokohama, Kanagawa 226-8503, Japan*

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Chiral phonons have nonzero polarization and can be observed only via a selective coupling with valley electrons and circularly polarized photons. In such process, a new physical quantity, i.e., pseudoangular momentum (PAM), is required to meet the selection rule. However, phonon PAM was only studied in symmorphic systems with quantized integer values. In this work, we generalized the definition of PAM to systems with screw rotation symmetry, i.e., nonsymmorphic systems, which show distinct properties compared with the one in symmorphic systems: (i) PAM can be nonintegers and $\mathbf{q}$ dependent but still be an observable quantity by experiments; (ii) phonon PAM and phonon angular momentum are not equal due the special Wyckoff position in the real space, in contrast to the previous works where most of the atoms are at rotation-invariant Wyckoff positions. Such new definition and discoveries can help us to obtain chiral phonons in a broader class of systems with a nonzero group velocity and to convey information like chirality and angular momentum in solids as expected. Materials are also offered to understand the new definition and for further experimental detection.

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Introduction. Chirality has been widely studied in fundamental physics since it reveals symmetry breaking of (quasi-)particles and governs many unconventional phenomena, such as chiral anomaly [1–3], unconventional Landau spectrum [4,5], Klein tunneling [6], nontrivial surface states [7–17], and so on. In recent years, chiral phonons draw much attention due to their contribution to many physical processes, such as electronic phase transition [18,19], giant thermal Hall effects in cuprate high- T_c superconductors [20,21], and valley polarization [22–29]. Chiral phonons can also transport information like chirality and angular momentum if they further couple with valley electrons and photons. However, most of them were only studied in two-dimensional (2D) systems at either the Brillouin zone (BZ) center or its boundaries [30–37], which means that they will have a vanishing group velocity due to the local extremal energy and reduces the efficiency of the information propagation. Thus, studies on chiral phonons in three-dimensional (3D) systems with a non-vanishing group velocity will help us to realize the chiral phonon-based quantum devices through materials.

Pseudoangular momentum (PAM) is proposed as a quantized physical quantity defined by the eigenvalue of a rotation operator [38] to offer a crucial selection rule to study chiral phonon absorption/emission in the intra-/intervalley electron scattering process via photon absorption. Although chiral phonons at high-symmetry points (HSPs) were observed by the helicity-resolved Raman scattering experiments in

transition-metal dichalcogenides and graphene by detecting the helicity flipping of the incident and emission photons [34,35,39–44], such a definition restricts the study of chiral phonons only in symmorphic space groups [45] and PAM was only studied in systems with rotation symmetry. In this work, we propose that PAM can be nonintegers and $\mathbf{q}$ dependent at any momenta with (screw) rotation symmetry, even in the nonsymmorphic systems (except for No. 7 and No. 9, which only have a glide mirror symmetry), which will be much easier to obtain chiral phonons with nonzero group velocity to convey information along expected directions. In most cases studied in previous works, all the atoms are C_n -rotation invariant Wyckoff positions (WPs), which we call case I, and the possible patterns of atomic motions are limited and simple. An atom at a C_n -rotation invariant position means that the position of the atom will be invariant under the C_n rotation operator. Other cases are classified into case II, and screw-symmetric systems always belong to this case II, having richer varieties than case I.

Chiral phonons and pseudoangular momentum. The chirality of phonons can be understood from the phonon polarization point of view, and regarded as one of the pieces of information carried by phonons. In 2D materials, phonon circular polarization for each mode is along the out-of-plane direction, such as WSe₂ in Figs. 1(a) and 1(b), while in 3D materials it can point to any direction due to the interatomic coupling in all directions. Yet, we can still define the phonon polarization, i.e., phonon chirality, for a 3D system by the vibration trajectory of atoms in the real space: if the atom vibrates in a counterclockwise circular/elliptical trajectory on a 2D plane, it will carry a counterclockwise chirality labeled by “+,” as shown in Fig. 1(e); otherwise, it will carry a clockwise chirality labeled by “−,” as shown in Fig. 1(f).

FIG. 1. (a), (b) Phase change of the phonon nonlocal part at K ($\frac{1}{3}, \frac{1}{3}$) under $\hat{C}_3$ for 2D material WSe_2 . $l^o = -1$ for the W atom in (a), while $l^o = +1$ for the Se atom in (b). (c), (d) Side and top views of the 3D material Te with screw rotation symmetry $\hat{C}_{3z}$. (d) Phase change of the phonon nonlocal part for Te at $(\frac{1}{3}, \frac{1}{3}, q_z)$ under $\hat{C}_{3z}$, which corresponds to $l^o = +q_z$. (e), (f) Schematic figures for two different vibration trajectories of the third and first modes for Te along the K - H high-symmetry line ($\frac{1}{3}, \frac{1}{3}, \frac{1}{4}$), which have an intracell part of pseudangular momentum with $l^s = +1, -1$, respectively. (g) Phonon spectra and BZ of Te.

As a selection rule for the optical transition process, PAM offers an additional conservation condition besides the crystal momentum conservation and energy conservation in the valley-dependent optoelectronics:

$$l_e^v + m = l_e^c + l_{ph}, \quad (1)$$

$$\hbar \mathbf{k}_e^v = \hbar \mathbf{k}_e^c + \hbar \mathbf{q}_{ph}, \quad (2)$$

$$E_e^v + E_{\text{photon}} = E_e^c + E_{ph}, \quad (3)$$

where $l_e^{v/c}$ is the electron PAM for the valence/conduction band, l_{ph} is the PAM for a phonon, $m = \pm 1$ represent the right/left-circularly polarized light, $\mathbf{k}_e$ and $\mathbf{q}_{ph}$ are the crystal momentum for electrons and phonons, and $E_{e/\text{photon}/ph}$ is the energy for electrons/photons/phonons.

The PAM was only studied in symmorphic space groups, in terms of the eigenvalue of n -fold rotation symmetry:

$$\hat{C}_n u_{\mathbf{q}} = e^{(-2\pi i/n)l_{ph,\mathbf{q}}} u_{\mathbf{q}}, \quad (4)$$

where $\hat{C}_n$ is the n -fold rotation operator, $u_{\mathbf{q}}$ is the phonon Bloch wave function, and $l_{ph,\mathbf{q}}$ is the phonon PAM at $\mathbf{q}$ with discrete values of $\{0, 1, \dots, n-1\} \bmod n$. We propose that PAM is still well defined and $\mathbf{q}$ dependent ($\mathbf{k}$ dependent for electrons) for nonsymmorphic systems with screw rotation symmetry via Eq. (4) with $\hat{C}_n$ replaced by the screw rotation

operator $\hat{C}_n'$. The definition can be extended for any (quasi-) particles at any momenta in solids preserving (screw) rotation symmetry.

Definition for intercell and intracell parts of PAM. Since the phonon Bloch wave function $u_{\mathbf{q}}$ has both a nonlocal contribution (Bloch-phase part l^o , which is also called intercell/orbital part in the previous works) from the Bloch phase factor $e^{i\mathbf{R}_l \cdot \mathbf{q}}$ ($\mathbf{R}_l$ is the atomic position) and a local contribution (self-rotation part l^s , which is also called intracell/spin part) from relative vibrations between sublattices, phonon PAM can be decomposed into $l_{ph} = l^s + l^o$ [46]. Like the total PAM, l^o and l^s should be also defined along the (screw) rotation axis direction. We find that motions of atoms for each phonon mode are totally different between two classes of systems, such as whether all the atoms are (screw) rotation-preserved sites (case I) or not (case II). In case I, all of the atoms are at the rotation-invariant WPs, and the value of l^o is uniquely determined by the WPs and the wave vector $\mathbf{q}$ (see the Supplemental Material for details [47]). We show that the possible combination of l^o and l^s is limited, because $l_{ph} = l^s + l^o$ is common for all the atoms. PAM equals AM when $\mathbf{q} = 0$ in case I. But in case II and some $\mathbf{q} \neq 0$ cases in case I, PAM does not equal AM. Cases in the previous works which belong to case I are among the list in the Supplemental Material [47], but such discussions in case II have not been done before. Thus, we will focus on case II in the following, and use Te as an example to show how to obtain l^o and l^s . It is worth mentioning that case II is the only possibility for nonsymmorphic systems with screw rotation symmetry.

The intercell/orbital part of PAM, i.e., l^o , is the phase difference of the Bloch phase factor between atoms in different sublattices related by (screw) rotation symmetry, and it depends on the phonon momentum $\mathbf{q}$ and the WP $\mathbf{R}_l$ in the real space. l^o is a quantized integer for symmorphic systems, while it becomes a $\mathbf{q}$ -dependent quantity for systems with screw rotation symmetry due to the fractional translation. For example, Figs. 1(a) and 1(b) show the phases of the phonon nonlocal part $e^{i\mathbf{R}_l \cdot \mathbf{q}}$ at $\mathbf{q} = K$ ($\frac{1}{3}, \frac{1}{3}$) for W and Se in WSe_2 , which belongs to case I. We note that all the coordinates in the real and reciprocal space are expressed in the basis of the primitive lattice vectors shown in Figs. 1(a) and 1(c). We take the $\hat{C}_3$ center to be at the center of the hexagon, which is the WP 1a. The W atom and Se atom are at WPs 1b and 1c, respectively (see the Supplemental Material for details [47]). The tungsten atom has $l_W^o = -1$ from the phase difference among the three $\hat{C}_3$ -related atoms in Fig. 1(a). Likewise, the selenium atom has $l_{Se}^o = +1$ in Fig. 1(b). This leads to $l_{ph} = l_W^o + l_{Se}^s = l_{Se}^o + l_{Se}^s$, i.e., $l_{ph}^s = l_{Se}^s + 2 \bmod 3$, meaning that $(l_W^o, l_{Se}^s) = (1, -1), (0, 1), (-1, 0)$, which agree with the combinations in the *ab initio* calculations in Ref. [34]. On the other hand, case II is the only possibility in systems with screw rotation symmetry. For example, Figs. 1(c), 1(d) and 1(g) are the crystal and phonon band structure of Te, which has a nonsymmorphic crystal structure $P3_121$ (No. 152) with a threefold screw rotation $\{\hat{C}_{3z} = C_{3z}|(0, 0, \frac{1}{3})\}$. Figure 1(d) shows the phases of the phonon nonlocal part $e^{i\mathbf{R}_l \cdot \mathbf{q}}$ at $\mathbf{q} = (\frac{1}{3}, \frac{1}{3}, q_z)$, and the fractional atomic positions for the three $\hat{C}_{3z}$ -related tellurium atoms are $\mathbf{R}_1 = (x, x, 0)$, $\mathbf{R}_2 = (-x, 0, \frac{1}{3})$, and $\mathbf{R}_3 = (0, -x, \frac{2}{3})$. Thus, $(e^{-i(2\pi/3)l^o})^3 = e^{-i\mathbf{q} \cdot \mathbf{R}_0} = e^{-2\pi i q_z}$,

i.e., $l^o = q_z$ for all the phonon modes in Te according to the phase difference, where $\mathbf{R}_0$ is the unit lattice vector along the c axis.

The intracell/spin part of PAM, l^s , which is a local contribution from the sublattice relative vibration, depends on the in-plane atomic vibration. In case I, $l^s = 1$ and $l^s = -1$ correspond to counterclockwise and clockwise rotations within the xy plane, and when $l^o = 0$, l^s can directly correspond to the AM. Since the possible situations for case I are very limited due to the symmetry restriction in both real and reciprocal space, we list all the possibilities in the Supplemental Material [47]. However, it will become much more complicated in case II, where l^s represents relative motions between the C_n -related atoms, instead of the atomic motion. In other words, l^s is the real space motion of a cluster, which is composed of all the C_n -related atoms. Thus, we need to calculate the phase difference between atoms related by screw rotation symmetry in nonsymmorphic systems. For example, Figs. 1(e) and 1(f) show the vibration trajectories of the third and first phonon bands for two $\hat{C}_3$ -related tellurium atoms at the middle point of the K - H line $(\frac{1}{3}, \frac{1}{3}, \frac{1}{4})$, respectively, where both of those tellurium atoms have a counterclockwise polarization. In Fig. 1(e), Te'_1 , which is obtained from Te_1 through the screw rotation $\hat{C}'_3$, has a $\frac{2\pi}{3}$ phase advancing to Te_2 and therefore this case corresponds to $l^s = +1$ for Te.

Distinct features of PAM in case II with screw rotation symmetry. We note that the definition of PAM in nonsymmorphic systems is not just an extension of that in symmorphic systems, but has unique features: (i) The noninteger nature of l_{ph} is from the fractional-translation part of the screw rotation symmetry, which is unique for nonsymmorphic systems. Namely, the phase difference between sublattices under screw rotation symmetry leads to a noninteger value of l^o . (ii) We only need to consider the atomic position components along the screw rotation axis when calculating l^o , which means that the atomic positions in plane will make no influence on l^o . (iii) Case II which we propose is the only possibility for nonsymmorphic systems with screw rotation symmetry. In the following, we will use nonsymmorphic crystal Te as an example to show those distinct features.

Electronic band structure of Te. Figure 3(a) is the spinful electronic band structure of Te ($P3_121$) with experimental lattice constants [48], and it shows that Te is a narrow gap insulator with a direct gap about $\Delta = 0.3$ eV at $P(\frac{1}{3}, \frac{1}{3}, q)$ and $PA(-\frac{1}{3}, -\frac{1}{3}, -q)$ in the vicinity of H [49]. Thus, chiral phonons involved in the valley scattering will be also away from HSPs and take values of $l_{ph,q} = 1 + q_z, q_z, q_z - 1 \bmod 3$ with $q_z \in [-0.5, 0.5]$. The highest two valence bands have a very large spin splitting about 0.18 eV, while the lowest two conduction bands have a spin splitting about 30 meV. Moreover, the spin component is different for the valence band maximum (VBM) and the conduction band minimum (CBM) at two valleys P and PA , which forbids the intravalley transition of electrons at each valley. Thus, both the multivalley band structure and different spin components for the VBM and CBM bands will be advantageous to realize selective coupling between chiral phonons and valley electrons via photon absorption.

FIG. 2. (a) Phonon spectra of Te along the H - K - H $(\frac{1}{3}, \frac{1}{3}, q_z)$ direction in the first BZ, where $q_z \in [-0.5, 0.5]$. “+” and “−” represent the chirality and l^s is the intracell part of the phonon angular momentum for each mode. Both the phonon chirality and l^s will keep the same value for each continuous phonon band. Real space trajectories for (b) the first and (c) the third mode at $(\frac{1}{3}, \frac{1}{3}, \frac{1}{4})$ with $l^s = -1$ and $l^s = +1$, respectively. (d) The highest three phonon modes along the H - K - H $(\frac{1}{3}, \frac{1}{3}, w)$ direction crossing three extended BZs, where $l^s = 0$, $l^o = l_{ph} = w$, and $w \in [-1.5, 1.5]$.

PAM of Te. Figure 2(a) shows the chirality and l^s for each phonon band in the first BZ. l^s takes the values 0, ± 1 and keeps the same value for each continuous band, while $l^o = q_z \in [-0.5, 0.5]$ is a $\mathbf{q}$ -dependent quantity. Thus, $l_{ph} = l^s + l^o$, taking the values $q_z, q_z \pm 1$, is also a $\mathbf{q}$ -dependent quantity. Consequently, l^s changes by ± 1 when q_z increases across the BZ boundary at H due to the $\hat{C}'_3$ screw symmetry. Table I shows the results of l^o , l^s , and l_{ph} for all the phonon modes at $(\frac{1}{3}, \frac{1}{3}, \frac{1}{4})$, and l_{ph} can be calculated by two ways: (i) the summation of the intracell and intercell parts of PAM, i.e., $l_{ph} = l^o + l^s$; (ii) l_{ph} is calculated directly from the eigenvalue of $\hat{C}'_{3z}$.

Since nine phonon modes are well separated into three groups in Fig. 2(a), each group can be understood as a result of band folding from a single phonon band extending over three BZs, and the values of l^s , l^o , and l_{ph} can be

TABLE I. l^o , l^s , and l_{ph} for each phonon band at $(\frac{1}{3}, \frac{1}{3}, \frac{1}{4})$ in tellurium, which is the middle point of the K - H line.

Band order	l^s	l^o	l_{ph} calculated by $l^s + l^o$	l_{ph} calculated by eigenvalue of $\hat{C}'_{3z}$
No. 1, No. 6, No. 8	−1	0.25	−0.75	−0.75
No. 2, No. 4, No. 9	0	0.25	0.25	0.25
No. 3, No. 5, No. 7	+1	0.25	1.25	1.25

understood in an extended scheme over three BZs [50]. Figure 2(d) shows the top three phonon bands [No. 7, No. 8, and No. 9 in Fig. 2(a)] in the extended three BZs, where the horizontal axis is the momentum along the H - K direction with $(\frac{1}{3}, \frac{1}{3}, w)$ and $w \in [-1.5, 1.5]$. There is only one optical phonon band from the vibration of one tellurium in the extended BZ, which leads to a trivial value of $l^s = 0$, $l^o = w$, and $l_{ph} = l^s + l^o = w$. By noting that the wave number $w \in [-1.5, 1.5]$ along the z axis in the extended scheme is related to that in the reduced scheme, $q_z \in [-0.5, 0.5]$, via $q_z = w \pmod{1}$, the result in Fig. 2(a) immediately follows from Fig. 2(d). Figures 2(b) and 2(c) show the trajectories of two C'_{3z} -related atoms for the first and third phonon modes, which is consistent with the l^s calculation explained in Figs. 1(e) and 1(f).

We note that Te has another chiral crystal structure of $P3_221$ (No. 154) with $\{C'_{3z} = C_{3z}|(0, 0, -\frac{1}{3})\}$, which will have the same phonon band spectra as $P3_121$. However, l_{ph} , l^o , and l^s will have an additional sign compared to the ones in $P3_121$ due to the opposite fractional translation along the z direction. Furthermore, l_{ph} , l^o , and l^s can be defined in the same way if systems have higher symmetries and higher degenerated modes. In systems with n_m screw symmetry, l^o takes the value of $l^o = mq_z \pmod{n}$. Thus, when q_z is increased across the BZ at $q_z = \pm\frac{1}{2}$, l^s jumps by m .

Chiral phonon-involved valley scattering in Te. As discussed above, the spin conservation condition forbids the intravalley transition in Te but allows the intervalley one between the CBM and VBM. In such process, phonons will be involved in the conservation of the total crystal momentum, PAM, and energy:

$$\mathbf{q}_{ph} = \mathbf{k}_{e,P} - \mathbf{k}_{e,PA}, \quad (5)$$

$$l_{e,P}^v + m + l_{ph} = l_{e,PA}^v \pmod{3}, \quad (6)$$

$$E_{\text{photon}} = E_{ph} + \Delta, \quad (7)$$

where $m = \pm 1$ represent the right/left-circularly polarized light and Δ is the electronic band gap of Te. We note that m is also not a quantized value due to the fractional translation of the screw rotation. However, we take $m = \pm 1$ since the fractional translation of the crystal is negligible compared to the wavelength of the photon.

Figures 3(b) and 3(c) show the chiral phonon absorption and emission process, and we will take the former one as an example. Since the momentum difference for electrons in the intervalley transition is $\mathbf{q}_{ph} = \mathbf{k}_{e,P} - \mathbf{k}_{e,PA} = (\frac{2}{3}, \frac{2}{3}, 2q)$, phonons with momentum of $\mathbf{q}_{ph} = (\frac{2}{3}, \frac{2}{3}, 2q)$ will be involved in this process with possible PAM values of $l_{ph} = 2q$, $2q + 1$, and $2q - 1$. Furthermore, l_e can be obtained from the eigenvalue of $\hat{C}'_{3z}$ at valley P and PA by first-principles calculation, i.e., $l_{e,P}^v = q - \frac{1}{2}$, $l_{e,PA}^v = -q + \frac{1}{2}$, $l_{e,P}^c = q - \frac{3}{2}$, $l_{e,PA}^c = -q + \frac{3}{2}$. Thus, by following Eq. (6), a chiral phonon

FIG. 3. New selection rules for phonon-involved intervalley scattering in 3D nonsymmorphic crystal Te. (a) Spinful electronic band structure of Te with a direct gap at $P(\frac{1}{3}, \frac{1}{3}, q)$. (b) By absorbing a chiral phonon with $l_{ph} = 2q - 1$, electrons can be scattered between P and PA valleys under the left-circularly polarized light ($m = -1$). (c) By emitting a chiral phonon with $l_{ph} = 2q - 1$, electrons can be scattered between two valleys under the right-circularly polarized light ($m = +1$).

absorption process can be obtained with $l_{ph} = 2q - 1$ by a left-circularly polarized incident photon with an energy Δ , as shown in Fig. 3(b). (Electrons can be scattered between those two valleys by emitting a chiral phonon with $l_{ph} = 2q - 1$ and $m = +1$, and they can also be scattered between other valleys by absorbing/emitting chiral phonons with different PAM.)

Conclusion. In this work, we not only generalize the definition of PAM (including l^s and l^o) to nonsymmorphic space groups at arbitrary momenta with screw rotation symmetry, but also discuss the more unique features of PAM that the screw rotation-preserved systems have than the symmorphic ones, which is due to the special WPs in the real space. The redefinition not only makes us realize that PAM can be nonintegers and $\mathbf{q}$ dependent, but also helps us to obtain chiral phonons with a broader condition. For example, Na_3Sb is a direct gap semiconductor with large Fermi velocity for the valence band maximum along different directions in the vicinity of A , which has the same crystal structure with Dirac semimetal Na_3Bi [51]. Since Na_3Sb preserves $\hat{C}'_6$ symmetry ($P6_3/mmc$), noninteger chiral phonons with $\text{PAM} \in [-3, 3] \pmod{6}$ can be observed by experiments if one gradually modulates the energy of the incident light to excite the electrons along the A - Γ direction [47].

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