Supporting Information for "Chiral Phonon Diode Effect in Chiral Crystals"

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The parameters of helix-chain model

For the phonon system, we consider a simple harmonic oscillator model described by the Hamiltonian

$$\mathcal{H} = \frac{1}{2}p^T p + \frac{1}{2}u^T K u, \tag{1}$$

where u is a column vector of displacements from the equilibrium positions, multiplied by the square root of mass (the mass is taken to be the same for every site here); p is the conjugate momentum vector, and K is the force constant matrix.

Along x direction, we set the force constant matrix as $k_x = \begin{bmatrix} k_L & 0 & 0 \\ 0 & k_{T1} & 0 \\ 0 & 0 & k_{T2} \end{bmatrix}$. Here, k_L is the longitudinal force constant, k_{T1} and k_{T2} are the transverse force constants. Along y-axis, we define a rotation operator as $T(\varphi) = \begin{bmatrix} cos\varphi & 0 & sin\varphi \\ 0 & 1 & 0 \\ -sin\varphi & 0 & cos\varphi \end{bmatrix}$, the force constant matrix $k_{x2} = T(\varphi)k_x T(-\varphi)$ has an angle φ with the $x-y$ plane.

Along the z-axis, we define a rotation operator as $U(\theta) = \begin{bmatrix} cos\theta & -sin\theta & 0 \\ -sin\theta & cos\theta & 0 \\ 0 & 0 & 1 \end{bmatrix}$. For the right-handed helix model, the force constant matrixes between atoms 2 and 3, atoms 1 and 2, atoms 1 and 3 are

$$\begin{aligned} k_1 &= U(\pi)k_{x2}U(-\pi), \\ k_2 &= U(\pi/3)k_{x2}U(-\pi/3), \\ k_3 &= U(-\pi/3)k_{x2}U(\pi/3), \end{aligned} \tag{2}$$

respectively.

Consider the nearest neighbor interaction, we can write the force constant matrix as $K = \begin{bmatrix} \frac{k_2+k_3}{m_1} & \frac{-k_2}{\sqrt{m_1 m_2}} & \frac{-k_3}{\sqrt{m_1 m_3}} \\ \frac{-k_2}{\sqrt{m_1 m_2}} & \frac{k_1+k_2}{m_2} & \frac{-k_1}{\sqrt{m_2 m_3}} \\ \frac{-k_3}{\sqrt{m_1 m_3}} & \frac{-k_1}{\sqrt{m_2 m_3}} & \frac{k_1+k_3}{m_3} \end{bmatrix}$. Considering the phase factor λ between the unit cell and its two nearest neighbors $\lambda_1 = e^{-i\boldsymbol{k}_z c}$, $\lambda_2 = e^{i\boldsymbol{k}_z c}$ where c is the z-direction helix period. Thus the 9×9 dynamic matrix can be expressed as

$$D(\boldsymbol{k}) = \begin{bmatrix} \frac{k_2+k_3}{m_1} & \frac{-k_2}{\sqrt{m_1 m_2}} & \frac{-k_3}{\sqrt{m_1 m_3}}\lambda_1 \\ \frac{-k_2}{\sqrt{m_1 m_2}} & \frac{k_1+k_2}{m_2} & \frac{-k_1}{\sqrt{m_2 m_3}} \\ \frac{-k_3}{\sqrt{m_1 m_3}}\lambda_2 & \frac{-k_1}{\sqrt{m_2 m_3}} & \frac{k_1+k_3}{m_3} \end{bmatrix}. \tag{3}$$

For the chiral chain model, the phonon mode eigenvector ϵ is solved from the eigenvalue problem

$$D(\boldsymbol{k})\epsilon(\boldsymbol{k},\sigma) = \omega^2_{\boldsymbol{k},\sigma}\epsilon(\boldsymbol{k},\sigma) \tag{4}$$

and σ is the phonon branch label. In this model, we set $\varphi = \pi/3$, $k_L = 1.0, k_{T1} = 0.05, k_{T2} = 0.25$ and $m_1 = m_2 = m_3 = 1.0$.

First-principles calculations details of α-quartz (SiO_2)

We performed the first-principles calculations to study the phononic properties of the α-quartz material. The calculations were done using the Vienna *ab initio* simulation package (VASP).[1,2] The projector augmented wave method[3] was adopted and the kinetic energy cutoff was set to be 500 eV. The generalized gradient approximation (GGA) with the Perdew-Burke-Ernzerhof (PBE)[4] realization was adopted for the exchange-correlation potential. The structure is fully optimized with the energy and force convergence criteria of 10^{-6} eV and 10^{-2} eV/Å, respectively and the Brillouin zone was sampled with k-mesh of size $9 \times 9 \times 9$. The phonon spectrum was obtained by using the PHONOPY code,[5] based on the force

constants calculated by the VASP-DFPT method. The $2 \times 2 \times 2$ supercell was used with a $3 \times 3 \times 3$ k-mesh. The method of non-analytical term correction (NAC)[6] was applied to get the dynamical matrix. And the lattice parameters of α-quartz are $a = b = 4.95$Å, $c = 5.45$Å.

Opposite total angular momentum effect

In equilibrium, the total phonon angular momentum of the system can be expressed as[7]

$$J_{\mathrm{ph}}^{\hat{n}} = \frac{1}{V} \sum_{\boldsymbol{k},\sigma} \hbar s_{\sigma}^{\hat{n}}(\boldsymbol{k}) [f_0(\omega_{\sigma}(\boldsymbol{k})) + \frac{1}{2}], \ \hat{n} = x, y, z \tag{5}$$

$$s_{\sigma}^{\hat{n}}(\boldsymbol{k}) = \epsilon_{\sigma}^{\dagger}(\boldsymbol{k}) \hat{S}_{\hat{n}} \epsilon_{\sigma}(\boldsymbol{k}), \tag{6}$$

where $f_0(\omega_{\sigma}(\boldsymbol{k})) = 1/(e^{\hbar\omega_{\sigma}(\boldsymbol{k})/k_B T} - 1)$ is the Bose-Einstein distribution. $\hbar s_{\sigma}^{\hat{n}}(\boldsymbol{k})$ represents the $\hat{n}$-direction phonon angular momentum of branch σ at point $\boldsymbol{k}$. V is the volume of the sample and T is the temperature, $\omega_{\sigma}(\boldsymbol{k})$ is the frequency corresponding to each phonon mode.

The number of left-handed phonons and right-handed phonons in the system are the same, which results in the total phonon angular momentum of the system being zero. After applying a temperature gradient, the phonon angular momentum of the system can be non-zero. By the Boltzmann transport theory, the form of the distribution function is

$$f_{\sigma,\boldsymbol{k}} = f_0(\omega_{\sigma}(\boldsymbol{k})) - \tau v_{\sigma,\hat{n}}(\boldsymbol{k}) \frac{\partial f_0}{\partial T} \frac{\partial T}{\partial x_{\hat{n}}}. \tag{7}$$

τ is the phonon relaxation time, $v_{\sigma,\hat{n}}$ is the phonon group velocity and $x_{\hat{n}}$ is the $\hat{n}$-component of the position. By substituting Eq. 7 into Eq. 5, we can obtain the total phonon angular momentum per unit volume under the temperature gradient as[8]

$$J_{\rm ph}^{\hat{n}} = -\frac{\tau}{V}\sum_{\boldsymbol{k},\sigma}\hbar s_{\sigma}^{\hat{n}}(\boldsymbol{k})v_{\sigma,\hat{m}}(\boldsymbol{k})\frac{\partial f_0(\omega_\sigma(\boldsymbol{k}))}{\partial T}\frac{\partial T}{\partial x_{\hat{m}}} = \alpha_{\hat{n}\hat{m}}\frac{\partial T}{\partial x_{\hat{m}}} \tag{8}$$

where $\alpha_{\hat{n}\hat{m}}$ denotes a response tensor. When the same z-direction temperature gradient is applied to the right-handed and left-handed lattices, they will produce opposite net phonon angular momentum due to the opposite distribution of chiral phonons. Because of the conservation of angular momentum, the rigid-body rotation angular momentum of the crystal needs to cancel out the net phonon angular momentum. If the system can rotate freely, the lattices with opposite chirality rotate in the opposite direction.

For the right-handed α-quartz material, the response tensor is $\alpha = \begin{pmatrix} \alpha_{xx} & 0 & 0 \\ 0 & \alpha_{yy} & 0 \\ 0 & 0 & \alpha_{zz} \end{pmatrix}$. At $T = 300\,\mathrm{K}$, the phonon angular momentum response tensor element $\alpha_{xx} = \alpha_{yy} = -6.5 \times 10^{-6} \times [\tau/(1\mathrm{s})]$ J s m^{-2}K^{-1} and $\alpha_{zz} = 4.1 \times 10^{-6} \times [\tau/(1\mathrm{s})]$ J s m^{-2}K^{-1} with relaxation time τ. For the left-handed α-quartz, it has the P3$_2$21 space group (154) and the response tensor element is opposite to the right-handed one.

Next, we estimate the angular velocity of rigid body rotation. Take a cylinder with radius r and height h, assuming that the phonon relaxation time is 10 ps. The z-direction temperature difference between the top and the bottom surfaces of the sample is represented by ΔT. The angular momentum of the rigid body rotation is expressed as $J_{\rm rotation}\pi r^2 h = I\omega$, where $I = \frac{1}{2}Mr^2$ is the momentum of inertia and M is the total mass of the sample. The angular velocity of the rigid-body rotation as

$$\omega = \frac{-J_{\rm ph}^z \pi r^2 h}{I} = \frac{-J_{\rm ph}^z}{\frac{1}{2}\rho r^2} \sim \frac{\Delta T/(1\mathrm{K})}{hr^2/(1\mathrm{m})^3} \times 10^{-20}\ \mathrm{s}^{-1}. \tag{9}$$

Assuming $\Delta T = 10$ K, when $h = 100\ \mu$m and $r = 10\ \mu$m, the angular velocity ω of rigid body rotation is estimated as $\omega \sim 10^{-5}$ s^{-1}. Crystals with opposite chirality rotate in opposite directions under the same temperature gradient.

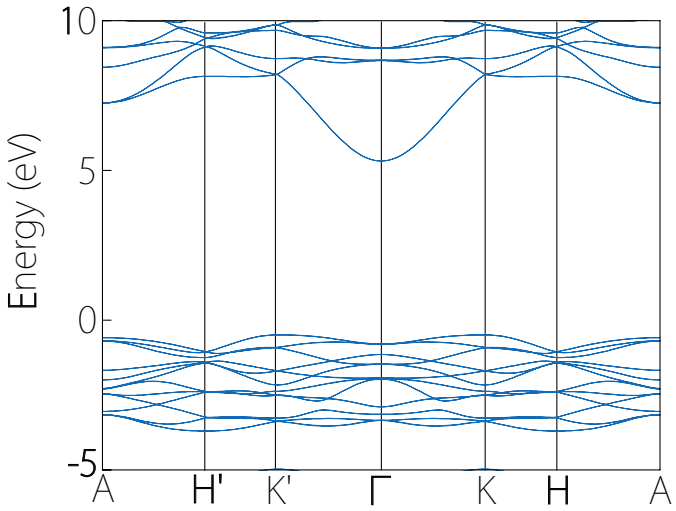

Figure 1: Electronic band structure of α-quartz.

Electronic band structure of α-quartz

Fig. 1 shows the electronic band structure of α-quartz, one can clear see that it is an insulator with a band gap of more than 5.8 eV. When irradiating circularly polarized light that responds to the energy of phonons, it can't stimulate the transition of electrons.

PAM of the chiral Te lattice

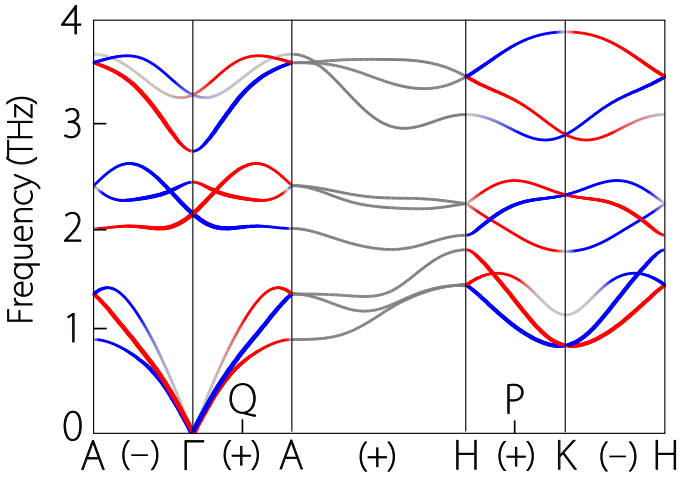

Figure 2: Phonon spectrum of the right-handed Te. $+/-$ on the path indicate the $k_z > 0$ / $k_z < 0$ part. We marked the phonon chirality. Red/blue color represents right/left-handed phonons. The phonon PAMs at the Q and P points are shown in the tables.

Table 1: Properties of the phonon modes at the Q point of the right-handed Te lattice. n labels the nine branches. $E_{\rm ph}$ is the phonon energy. $\pm$ denotes the mode has right-/left-handed chirality. $\ell_{\rm ph}$ is the phonon PAM.

n	$E_{\rm ph}$ (cm^{-1})	chirality	$\ell_{\rm ph}$
1	23.1	+	1
2	26.5	−	-1
3	35.9	+	0
4	67.3	−	1
5	76.8	+	0
6	86.8	+	-1
7	110.7	−	0
8	116.0	−	1
9	122.3	+	-1

Table 2: Properties of the phonon modes at the P point of the right-handed Te lattice. n labels the nine branches. $E_{\rm ph}$ is the phonon energy. $\pm$ denotes the mode has right-/left-handed chirality. $\ell_{\rm ph}$ is the phonon PAM.

n	$E_{\rm ph}$ (cm^{-1})	chirality	$\ell_{\rm ph}$
1	34.6	−	-1
2	42.9	+	1
3	50.6	+	0
4	64.7	+	0
5	74.8	−	1
6	82.5	+	-1
7	98.6	−	1
8	108.5	+	-1
9	127.0	−	0

Fig. 2 shows the phonon spectrum of the right-handed Te lattice. The calculated phonon chirality and PAM of the right-handed Te lattice are shown in the Table 1 and Table 2.

The calculations of phonon spectrum are done using the Quantum Espresso package.[9] The local density approximation (LDA) is adopted for the exchange-correlation functional. The kinetic energy cutoff is set to be 60 Ry. The dynamical matrices are obtained with a $4 \times 4 \times 4$ k-mesh, within the framework of density functional perturbation theory (DFPT). And the lattice parameters of two chiral Te lattices are $a = 4.51$Å, $c = 5.96$Å.

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