Truly chiral phonons in $\alpha$-HgS observed by circularly polarised Raman spectroscopy

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Chirality is a manifestation of the asymmetry inherent in nature. Chirality has been defined as the symmetry breaking of the parity of static objects$^{1}$, and the definition was extended to dynamic motion such that true and false chiralities were distinguished$^{2}$. Recently, rotating, yet not propagating, atomic motions were predicted and observed in two-dimensional materials, and they were referred to as ‘chiral phonons’$^{3–5}$. A natural development would be the discovery of truly chiral phonons that propagate while rotating in three-dimensional materials. Here, we used circularly polarised Raman scattering and first-principles calculations to identify truly chiral phonons in chiral bulk crystals. This approach enabled us to determine the chirality of a crystal in a noncontact and nondestructive manner. In addition, we demonstrated that the law of the conservation of pseudoangular momentum holds between circularly polarised photons and chiral phonons. These findings are foreseen to pave the way for transferring the pseudoangular momentum from photons to electron spins via the
propagating chiral phonons in opto-phononic-spintronic devices.

Chirality, the breaking of reflection and inversion symmetries, appears at all levels of nature and has been studied in many fields of research. Barron extended the concept of chirality to the dynamic regime as classified into true and false chiralities. True chirality is demonstrated by systems that exist in two different enantiomeric states that are interconverted by spatial inversion ($P$), rather than by time inversion ($T$) combined with appropriate spatial rotation ($R$). This concept contrasts with false chirality, where the two states are interconverted by $P$ and by $RT$; for example, the magneto-optical Faraday rotation.

Chirality was further extended to the dynamics of quasi-particles. Possible band-structure shapes have been classified in terms of pseudomomentum and pseudoangular momentum (PAM), which originate from the phase factors acquired by discrete translation and rotation symmetry operations on wave functions, respectively. In crystals, they are different from the momentum and angular momentum (AM), which are continuous linear and circular motions of an object in real space, respectively. Recently, circularly rotating, yet not propagating, atomic motion with nonzero PAM and AM at high symmetry points in the Brillouin zone in monolayer hexagonal lattices was theoretically predicted and named ‘chiral phonons’. This prediction was experimentally verified in the transition metal dichalcogenide WSe$_2$ via transient infrared (IR) spectroscopy.

Employing Barron’s true chirality, one can extend the concept of a chiral phonon to ‘truly chiral phonons’, which have two enantiomeric chiral modes that propagate while circularly rotating in three-dimensional materials with opposite signs of PAM and AM. These modes should be interconverted by $P$, but not by $RT$. The observation of truly chiral phonons would require the use of chiral materials, as has been proposed theoretically. Several Raman experiments designed to identify phonon symmetry in chiral crystals were conducted. Frequency-degenerated phonon modes were observed to undergo splitting at the centre of the Brillouin zone ($\Gamma$ doublets) in $\alpha$-quartz and trigonal tellurium. Moreover, theoretical work on phonon band dispersion has been reported on $\alpha$-HgS (cinnabar), where the splitting was attributed to the linear wave-vector
shifts caused by chirality\textsuperscript{[15]}. Nonetheless, previous studies have mostly focused on examining the phonon symmetry without considering PAM and AM.

Here, we used circularly polarised Raman spectroscopy (see Methods) to observe the phonon modes and their splitting in chiral crystals of $\alpha$-HgS. Moreover, we performed first-principles calculations (see Methods) to compute the dispersion, PAM, and AM of phonons in $\alpha$-HgS. A comparison between the experiment and calculation shows that the split phonons were indeed truly chiral phonons. We also confirmed that the conservation law of PAM holds in circularly polarised Raman scattering by taking into account the three-fold rotational symmetry.

Trigonal $\alpha$-HgS crystals contain two enantiomeric cinnabar structures with right- and left-handed helices (R- and L-HgS), which belong to the space groups $P3_121$ and $P3_221$, respectively\textsuperscript{[17]}. This crystal has a three-fold helical axis along the $c$-direction (Fig. 1(a)), with large optical activity\textsuperscript{[18]}. Our samples were single crystals of naturally grown $\alpha$-HgS with lateral dimensions of 3 mm. We selected a $c$-axis-oriented as-grown surface of R-HgS, which was confirmed by X-ray analysis.

The phonon modes at the centre of the Brillouin zone (Γ point) of $\alpha$-HgS are classified into two $\Gamma_{1}^{(1,2)}$ singlets (Raman active), three $\Gamma_{2}^{(1-3)}$ singlets (IR active), and five $\Gamma_{3}^{(1-5)}$ doublets (Raman and IR active). Our measurement led to the observation of the $\Gamma_{1}^{(1,2)}$ and $\Gamma_{3}^{(2-5)}$ doublets, which were assigned on the basis of the selection rule\textsuperscript{[19]} for the four incident/scattered polarisation configurations (RR, LL, RL, and LR), where $I_{RR} : I_{LL} : I_{RL} : I_{LR} = 1 : 1 : 0 : 0$ and $0 : 0 : 1 : 1$ for the $\Gamma_{1}$ and $\Gamma_{3}$ modes, respectively (Extended Data Fig. 1). The transverse optical (TO) phonons of the $\Gamma_{3}$ mode were generated because the incident and scattered light were propagated along the $c$-axis. A weak signal was detected for the longitudinal optical (LO) phonon at $\sim 145$ cm$^{-1}$, presumably due to misalignment of the experimental setup.

The phonon frequencies obtained by the Raman experiment are compared with the results of the first-principles calculations and with previous results\textsuperscript{[10,16]} in Extended Data Table 1.
The properties of phonons are discussed here on the basis of our first-principles calculations. Figure 3(a) displays the phonon dispersion curve calculated along the path $\Delta$ from $\Gamma$ to A point, which is parallel to the $c$-axis [see Fig. 1(b)]. Linear splittings of $\Gamma_3^{(1-5)}$ modes appear around the $\Gamma$ point, which is consistent with previous calculations\cite{16,20}. From the conservation law of pseudomomentum, the wavenumber $k$ of phonons observed by backscattering Raman spectroscopy is expressed as $k = 4\pi n/\lambda$, where $\lambda$ is the wavelength of the incident or scattered light, $n = 2.77$ is the absolute value of the complex refractive index of $\alpha$-HgS\cite{21}. The value of $k$ was calculated to be $\sim 1/80$ of the wavenumber $\pi/c$ at the A point. We denoted this specific point on the path $\Delta$ as ‘N’ in Fig. 3(a). The experimental splitting values of the $\Gamma_3$ modes at this point (Stokes scattering) are almost consistent with the calculations, as indicated in Extended Data Table 2. Therefore, we attribute the observed splittings in Fig. 2 to the linear $k$ shifts at the N point.
To understand the selection rule of the incident/scattered polarisation configurations on the Raman spectra in Fig. 2, we calculated the PAM of phonons in R-HgS (see Methods). As shown in Fig. 3(a), the $\Gamma_3$ doublets have spin PAM $l_{\text{PAM}}^s = \pm 1$, whereas the $\Gamma_1$ and $\Gamma_2$ singlets have $l_{\text{PAM}}^s = 0$. The phonon bands with $l_{\text{PAM}}^s = -1$ and $l_{\text{PAM}}^s = 0$ intersect at the A point. These are the common features of the $3_1$ (right-handed) helix $^{7,9,22}$. From Figs. 2(e–h) and 3(a), $\Gamma_3^{(2–5)}$ phonons with $l_{\text{PAM}}^s = +1$ and $-1$ were observed in the LR and RL configurations, respectively (Extended Data Table 2). $\Gamma_1$ phonons with $l_{\text{PAM}}^s = 0$ were observed in the RR and LL configurations. Right- and left-handed circularly polarised light possess PAM of $\sigma = +1$ and $-1$, respectively $^{23}$. Therefore, we confirmed a conservation law to exist between the spin PAM of phonons and the PAM of the incident/scattered photons in the Stokes spectra as

$$\sigma_s - \sigma_i = -l_{\text{PAM}}^s + 3p,$$ (1)

where $\sigma_s$ and $\sigma_i$ represent the PAM of the scattered and incident photons, respectively. In addition, $p = 0$ and $\pm 1$ for the $\Gamma_1$ and $\Gamma_3$ modes, respectively. The factor $3p$ on the right-hand side of equation (1) can be understood by considering the three-fold rotational symmetry of $\alpha$-HgS and the Umklapp process in Raman scattering $^{7,24–27}$.

In crystals with discrete rotational symmetry, the PAM and AM of phonons should be distinguished. The AM of phonons arises from the circular vibration of atoms in the real space, as shown in Figs. 1(c–f) [See also Supplementary Movies 1 and 2]. The AM of phonons along the $c$-axis is displayed in Fig. 3(b) along with $\Gamma$ to A point (see Methods). In the vicinity of the $\Gamma$ point, the $\Gamma_3$ modes have clockwise and counter clockwise rotations, corresponding to the positive and negative AM of phonons, respectively. This is a clear manifestation of the existence of chiral phonons. The $\Gamma_1$ modes have linear vibrations with zero AM of phonons. The signs of AM at the N point are listed in Extended Data Table 2, where the spin PAM and AM of phonons do not correspond with each other. This indicates that, contrary to PAM, the AM of phonons is not conserved during the Raman process.

We also calculated the PAM and AM of phonons in L-HgS (Extended Data Fig. 2). The signs
of spin PAM and AM are reversed with respect to Fig. 3, that is, \( l_{PAM}^s(RH, k, j) = -l_{PAM}^s(LH, k, j) \) and \( l_{AM}^s(RH, k, j) = -l_{AM}^s(LH, k, j) \), where RH and LH denote crystals with right- and left-handed helices, respectively. This means that the splitting of the \( \Gamma_3 \) modes arises from chirality. From the first-principles calculation, the phonon eigenvector at the N point \( u(RH, k, j) \) and \( u(LH, k, j) \) are converted by \( P \), \( T \), and \( C_2 \) as \( P u(RH, k, j) = u(LH, -k, j) \), \( C_2 T u(RH, k, j) = C_2 u(RH, -k, j) = u(RH, k, j) \), where \( C_2 \) is a two-fold rotation around the \( a \), \( b \), or \( a + b \) direction. These relations satisfy the definition of true chirality with two different enantiomeric phonon modes: \( u(RH, k, j) \) and \( u(LH, -k, j) \).

We also recorded the Raman spectra of an \( \alpha \)-HgS crystal for which the handedness (chirality) of the measured region was unknown because the size of the chiral domain was smaller than the experimental resolution (\( \sim 1 \) mm) of our X-ray diffractometer. Contrary to the X-ray analysis, our Raman experiment with a spatial resolution of a few \( \mu \)m succeeded in observing the splitting of the \( \Gamma_3 \) modes (Extended Data Fig. 3), and the directions of the splittings were opposite to the results in Fig. 2. Considering the data in Extended Data Table 3, the chirality of the sample was found to be left-handed.

The space group analysis enabled us to additionally assign an irreducible representation of the phonon dispersion curve in R-HgS. On path \( \Delta \) from \( \Gamma \) to A, three irreducible representations are known to exist: \( \Delta_1 \), \( \Delta_2 \), and \( \Delta_3 \). The labels of the irreducible representations are based on the convention in Ref. 28. The \( \Gamma_1 \) and \( \Gamma_2 \) modes at the \( \Gamma \) point change to the \( \Delta_1 \) mode, whereas the \( \Gamma_3 \) modes split into \( \Delta_2 \) and \( \Delta_3 \) modes in accordance with the compatibility relations. In the Supplementary Information, we present a derivation of the Raman tensors that correspond to the \( \Delta_2 \) and \( \Delta_3 \) modes. It follows that the Raman intensities of the \( \Delta_2 \) and \( \Delta_3 \) modes are \( I_{RR} : I_{LL} : I_{RL} : I_{LR} = 0 : 0 : 1 : 0 \) and \( 0 : 0 : 0 : 1 \), respectively. This is consistent with the results of the Raman experiments. In other words, spin PAM \( l_{PAM}^s(RH) = 0, +1, -1 \) correspond to \( \Delta_1, \Delta_2, \Delta_3 \), respectively, because the phase change by three-fold rotation is the index of each irreducible representation. For L-HgS, \( l_{PAM}^s(LH) = 0, +1, -1 \) correspond to \( \Delta_1, \Delta_3, \Delta_2 \), respectively28.
Next, we discuss the propagation of the chiral phonons. From Fig. 3, the group velocities of the $\Gamma_3^{(1-5)}$ modes at the N point are calculated as $\sim \pm 0.4-2 \times 10^3$ m/s, which is comparable to the sound velocity of acoustic phonons. Note that the nonreciprocal propagation of chiral phonons can be controlled by the PAM of the photon. This implies the possibility of transferring the PAM from photons to electron spins via the propagating chiral phonons in opto-phononic-spintronic devices.

Weyl phonons can exist widely in chiral crystals, which have been predicted by the first-principles calculations and verified by inelastic X-ray scattering. We note that circularly polarized Raman spectroscopy presented in this paper may open the possibility to detect phonons carrying nonzero Chern number through measuring the phonon PAM, instead of detecting the eigenvalue and eigenvectors of the topological bands.

We observed chiral phonons in a three-dimensional chiral system using circularly polarized Raman spectroscopy and first-principles calculations. The chiral phonons were labelled with spin PAM of \{+1, −1\} corresponding to $\{\Delta_2, \Delta_3\}$ and $\{\Delta_3, \Delta_2\}$ for R- and L-HgS, respectively, with opposite group velocities of $\sim 1$ km/s. The parity and time-reversal symmetries of the phonons satisfy the definition of truly chiral phonons, which propagate while rotating along the $c$-axis. This is distinct from the chiral phonons observed in two-dimensional hexagonal systems. Our work also provides an optical method to identify the handedness of chiral materials using PAM and we demonstrated that the spatial imaging of chiral domains can be achieved in a noncontact and nondestructive manner.

**Data availability** The data that support the findings of this study are available from the corresponding author upon reasonable request.

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**Author contributions**

T.S. and J.K. conceived the study. K.I. and H.M. performed the Raman experiments. Y.K., Y.T., and S.I. conducted the X-ray analyses. K.I., H.M., T.Z., and S.M. performed the first-principles calculations. K.I., M.H., J.K., and T.S. wrote the manuscript. All authors discussed the results and commented on the manuscript.
Competing interests

The authors declare no competing interests.

Methods

Raman scattering. We used a custom-built Raman spectroscopy system equipped with a monochromator, charge-coupled device (CCD) camera, and optical elements. Details of the setup can be found elsewhere. Excitation light was generated by a diode laser with a wavelength of $\lambda = 785$ nm. The incident and scattered light were propagated along the $c$-axis of the crystal. All the measurements were performed at room temperature.

First-principles calculation. First-principles calculations were performed using the ABINIT package which implements the density functional perturbation theory (DFPT). An anaddb was used to obtain the phonon dispersion curve. For the calculation of phonon dispersion, the exchange-correlation energy of electrons is described as the generalized gradient approximation (GGA) with the norm-conserving pseudopotentials, and spin-orbit coupling was not considered. We set the kinetic-energy cutoff to 60 Ha, which was necessary to obtain a well-converged result, and the grids of the wave vector $k$ to describe the phonon dispersion curve. In addition, we increased the precision of the $k$ grids to 8 x 8 x 8 to obtain the converged phonon frequency at the $\Gamma$ point. The lattice parameters of $\alpha$-HgS were used as follows: volume of the cell $V_0 = 160.02 \, \text{Å}^3$, and the length of the cell is $a = 4.11 \, \text{Å}$, $b = 4.11 \, \text{Å}$, and $c = 9.40 \, \text{Å}$, the angle of cell $\alpha = 90^\circ$, $\beta = 90^\circ$, and $\gamma = 120^\circ$. The essential part of the ABINIT results was confirmed using the VASP package.

Calculation of PAM. In a material with three-fold rotational symmetry, PAM has been defined as follows

$$\{C_3 \mid 0\} \, \mathbf{u}(\mathbf{k}) = \exp \left[ -i \frac{2\pi}{3} l_{\text{PAM}}(\mathbf{k}) \right] \, \mathbf{u}(\mathbf{k}),$$

where $l_{\text{PAM}}(\mathbf{k})$ is the PAM of phonons, $\mathbf{u}(\mathbf{k})$ is the displacement vector, and $\{C_3 \mid 0\}$ is the three-
fold rotation around the c-axis. However, α-HgS has a three-fold helical rotational symmetry instead of a three-fold rotational symmetry. Therefore, we defined the PAM of phonons in R- and L-HgS as follows:

\[
\{C_3 | c/3\} u(RH, k) = \exp\left[-i \frac{2\pi}{3} l_{PAM}(RH, k)\right] u(RH, k),
\]

\[
\{C_3 | 2c/3\} u(LH, k) = \exp\left[-i \frac{2\pi}{3} l_{PAM}(LH, k)\right] u(LH, k),
\]

where \(\{C_3 | c/3\}\) and \(\{C_3 | 2c/3\}\) are combinations of the three-fold rotation around the c-axis and the c/3 and 2c/3 translations along the c-axis, respectively.

The displacement vector of phonons can be expressed as follows

\[
u_\kappa(k, l, j) = m_\kappa^{-\frac{1}{2}} \epsilon_\kappa(k, j) \exp[i \{k \cdot R_l - \omega(k, j) t\}],
\]

where \(j\) is the number of phonon modes, \(m_\kappa\) is the mass of the \(\kappa\)-th atom in the \(l\)-th unit cell, \(\epsilon_\kappa(k, j)\) is the eigenvector of the dynamical matrix, and \(R_l\) is the position of the \(l\)-th unit cell.

The use of equation (5) indicates that the phase of the eigenvector consists of two parts: \(\epsilon(k)\) and \(\exp[i k \cdot R_l]\), both of which contribute to the calculation of the PAM. In the former, the phase difference of \(\epsilon(k)\) leads to the spin PAM \(l_{PAM}^s\), which is quantized as an integer. The latter factor, \(\exp[i k \cdot R_l]\), provides the orbital PAM \(l_{PAM}^o\), which is therefore equal to \(\frac{1}{2\pi} k \cdot c\) under a three-fold helical rotation. Consequently, for each wavenumber \(k\), the total PAM is

\[l_{PAM} = l_{PAM}^s + l_{PAM}^o.
\]

Here, we considered only the spin PAM in the phonon dispersion, as shown in Fig. 5(a).

**Calculation of AM.** The AM of phonons at wavenumber \(k\) of mode number \(j\) is defined as

\[l_{AM}(k, j) = (\epsilon(k, j)^T M \epsilon(k, j)) \hbar.
\]

where

\[
M = \begin{pmatrix}
0 & -i & 0 \\
i & 0 & 0 \\
0 & 0 & 0
\end{pmatrix} \otimes I_{n \times n},
\]

where \(n\) is the number of atoms in a unit cell and \(I_{n \times n}\) is a
unit matrix of $n \times n$. The eigenvector of the dynamical matrix $\epsilon(k, j)$ is normalised as:

$$
\epsilon(k, j)\epsilon(k, j)^\dagger = 1.
$$

(8)

The origin of spin and orbital PAMs. Here we provide an explicit form of the spin and orbital PAMs associated with the screw operation $\{C_3 | c/3\}$, where $c/3$ is the non-primitive translation. The atomic displacement amplitude associated with the phonon propagation may be generally written as

$$
U_k(r) = \epsilon(k) \exp(i k \cdot r).
$$

(9)

The vector field, $U_k(r)$, is then transformed in accordance with a general rule:

$$
\{C_3 | c/3\} U_k(r) = [C_3\epsilon(k)] \exp(i k \cdot \{C_3 | c/3\}^{-1} r).
$$

(10)

Note that $\{C_3 | c/3\}^{-1} = \{C_3^{-1} | -C_3^{-1} c/3\}$, $k \cdot r$ is a scalar, and $C_3\epsilon(k) = \exp(-i\frac{2\pi}{3} m) \epsilon(k)$, we have

$$
\{C_3 | c/3\} U_k(r) = \exp\left[-i\frac{2\pi}{3} (l_{\text{PAM}}^s + l_{\text{PAM}}^o)\right] U_k(r)
$$

(11)

where the respective spin and orbital pseudoangular momenta are introduced by

$$
l_{\text{PAM}}^s = m,
$$

(12)

and

$$
l_{\text{PAM}}^o = -\frac{3}{2\pi} \{(C_3k) - k\} \cdot r + \frac{1}{2\pi} (C_3k) \cdot c,
$$

(13)

which yield the total PAM, $l_{\text{PAM}} = l_{\text{PAM}}^s + l_{\text{PAM}}^o$.

In this study, we consider path $\Delta$ from $\Gamma$ to $A$, where $C_2k = k$ denotes the modulo reciprocal lattice vectors. Thus, $l_{\text{PAM}}^o = \frac{1}{2\pi} k \cdot c$. The same symmetry properties apply to the photons propagating along the helical axis. Then, we obtain the selection rule (1), which includes only $l_{\text{PAM}}^s$, by cancelling out the $l_{\text{PAM}}^o$ terms because of the law of conservation of momentum.

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Figure 1: Chiral properties of $\alpha$-HgS. (a) Right-handed helical chain along the $c$-axis of R-HgS. Each Hg atom (red sphere) is bonded with two S atoms (blue spheres). Chemical bonds are denoted as green lines. (b) Brillouin zone of $\alpha$-HgS. (c) Schematic diagram of phonon dispersion from $\Gamma$ to A point. The red, blue, and black circles at the N point correspond to the phonon modes of panels (d), (e), and (f), respectively. Two-dimensional projections of schematic atomic motions in R-HgS at point N for (d) a $\Delta_2$ mode with $l_{\text{PAM}}^r = +1$, $l_{\text{AM}} > 0$, (e) a $\Delta_3$ mode with $l_{\text{PAM}}^r = -1$, $l_{\text{AM}} < 0$, and (f) a $\Delta_1$ mode with $l_{\text{PAM}}^r = 0$ and $l_{\text{AM}} \simeq 0$. Counter clockwise, clockwise circular, and linear motions indicate that $l_{\text{AM}}$ is positive, negative, and zero, respectively. The three-fold rotation symmetry operation generates phase factors of atomic motions, namely, PAM $l_{\text{PAM}}^r$. The phase difference at the position of each atom after the operation is shown.
Figure 2: **Raman spectra of R-HgS.** (a–d) Anti-Stokes and (e–h) Stokes spectra of the $\Gamma_3$ modes. The black and red lines denote the $\Gamma_3$ modes experimentally observed with RL- and LR-polarised configurations, respectively. The values of the $\Gamma_3$ doublet splittings are shown in each figure.
Figure 3: **Phonon dispersion curve of R-HgS along the right-handed helical axis.** (a) Black, red, and blue curves correspond to spin PAM $l_{PAM} = 0, +1, -1$, respectively. The wavenumbers at the N and A points are in the ratio of 1 : 80. (b) Phonon AM, denoted by the colour gradient. The red and blue curves correspond to the positive and negative AM, respectively.
Extended Data Fig. 1 | Full Raman spectra of R-HgS. Full Raman spectra were recorded at room temperature using various polarised configurations. The black, green, red, and blue spectra represent the LL-, RR-, LR-, and RL-polarised configurations, respectively. R and L represent the right- and left-handed helicities of circularly polarised light, respectively.
Extended Data Fig. 2 | Phonon dispersion curve of L-HgS along the left-handed helical axis. (a) The black, red, and blue curves correspond to spin PAM $l_{\text{PAM}}^s = 0, +1, -1$, respectively. (b) Phonon AM, denoted by the colour gradient. The red and blue curves correspond to the positive and negative AM, respectively.
Extended Data Fig. 3 | Raman spectra of α-HgS with unknown chirality. (a–d) Anti-Stokes and (e–h) Stokes spectra of the $\Gamma_3$ modes. The black and red lines denote the $\Gamma_3$ modes experimentally observed with RL- and LR-polarised configurations, respectively. R and L represent the right- and left-handed helicities of circularly polarised light, respectively. The values of the $\Gamma_3$-doublet splittings are shown in each figure.
Extended Data Table 1 | phonon frequencies in α-HgS. The experimental values in cm$^{-1}$ were obtained at room temperature. The calculated values in cm$^{-1}$ are at the $\Gamma$ point.

| Symmetry | Our experiments | Experiments$^{[10]}$ | Our calculations | Calculations$^{[15]}$ |
|----------|-----------------|---------------------|------------------|---------------------|
| $\Gamma_{2,LO}^{(1)}$ | Inactive | | 40.2 | 44.1 |
| $\Gamma_1^{(1)}$ | 42 | 45 | 41.0 | 39.1 |
| $\Gamma_{3,TO}^{(1)}$ | | | 43.5 | 42.0 |
| $\Gamma_{3,TO}^{(2)}$ | 85 | 88 | 84.4 | 83.1 |
| $\Gamma_{3,TO}^{(3)}$ | 103 | 106 | 114.4 | 121.4 |
| $\Gamma_{2,LO}^{(2)}$ | Inactive | | 149.4 | 159.6 |
| $\Gamma_1^{(2)}$ | 254 | 256 | 237.5 | 232.1 |
| $\Gamma_{3,TO}^{(4)}$ | 282 | 283 | 263.0 | 259.3 |
| $\Gamma_{3,TO}^{(5)}$ | 344 | 345 | 320.2 | 319.2 |
| $\Gamma_{2,LO}^{(3)}$ | Inactive | | 339.4 | 337.7 |
**Extended Data Table 2 | Chiral properties of phonons in R-HgS along the right-handed helical axis.** Owing to the roughness of the sample surface, multiple splitting measurements were conducted, with the deviation indicated by the error bars.

| Symmetry at Γ point | Experiment | Calculation | | | |
|---|---|---|---|---|---|
| | Config. | Frequency [cm$^{-1}$] | Splitting [cm$^{-1}$] | Symmetry at N point | Frequency [cm$^{-1}$] | Splitting [cm$^{-1}$] | $l_{PM}^3$ | $l_{AM}$ |
| $\Gamma_3^{(2)}$ | RL | 84.9 | 0.3 ± 0.01 | $\Delta_2^{(2)}$ | 84.2 | 0.4 | −1 | − |
| | LR | 85.3 | | $\Delta_3^{(2)}$ | 84.6 | | +1 | + |
| $\Gamma_3^{(3)}$ | LR | 103.2 | 0.3 ± 0.16 | $\Delta_2^{(3)}$ | 114.0 | 0.9 | +1 | − |
| | RL | 103.5 | | $\Delta_3^{(3)}$ | 114.9 | | −1 | + |
| $\Gamma_3^{(4)}$ | RL | 281.9 | 0.4 ± 0.08 | $\Delta_2^{(4)}$ | 262.7 | 0.7 | −1 | + |
| | LR | 282.3 | | $\Delta_3^{(4)}$ | 263.4 | | +1 | − |
| $\Gamma_3^{(5)}$ | LR | 343.5 | 0.3 ± 0.05 | $\Delta_2^{(5)}$ | 320.0 | 0.4 | +1 | + |
| | RL | 343.7 | | $\Delta_3^{(5)}$ | 320.4 | | −1 | − |
Extended Data Table 3 | Chiral properties of phonons in α-HgS with unknown chirality. The error bars take into account that multiple splitting measurements were conducted in the experiment, owing to the roughness of the sample surface. Calculations were performed for L-HgS.

| Symmetry at Γ point | Experiment | Calculation |
|---------------------|------------|-------------|
|                     | Config.    | Frequency [cm⁻¹] | Splitting [cm⁻¹] | Symmetry at N point | Frequency [cm⁻¹] | Splitting [cm⁻¹] | \(l^{\text{PAM}}\) | \(l^{\text{AM}}\) |
| \(\Gamma_3^{(2)}\) | LR         | 84.6         | 0.4 ± 0.04    | \(\Delta_3^{(2)}\) | 84.2         | 0.4          | +1             | +             |
|                    | RL         | 85.0         |               | \(\Delta_2^{(2)}\) | 84.6         | -1           | -              | -              |
| \(\Gamma_3^{(3)}\) | RL         | 102.9        | 0.4 ± 0.04    | \(\Delta_3^{(3)}\) | 114.0        | 0.9          | -1             | +             |
|                    | LR         | 103.3        | 0.4 ± 0.04    | \(\Delta_2^{(3)}\) | 114.9        | 1            | +1             | -              |
| \(\Gamma_3^{(4)}\) | LR         | 281.9        | 0.5 ± 0.08    | \(\Delta_3^{(4)}\) | 262.7        | 0.7          | +1             | -             |
|                    | RL         | 282.4        |               | \(\Delta_2^{(4)}\) | 263.4        | -1           | +1             | +             |
| \(\Gamma_3^{(5)}\) | RL         | 343.1        | 0.4 ± 0.03    | \(\Delta_2^{(5)}\) | 320.0        | 0.4          | -1             | -             |
|                    | LR         | 343.5        | 0.4 ± 0.03    | \(\Delta_3^{(5)}\) | 320.4        | 1            | +1             | +             |