

Chiral phonons in helical single crystal Te by circularly polarized Raman spectroscopy

Kyosuke Ishito¹, Huiling Mao¹, Kaya Kobayashi², Jun-ichiro Kishine³, Takuya Satoh¹

(¹Department of Physics, Tokyo Institute of Technology, Tokyo, 152-8551, Japan;

²Research Institute for Interdisciplinary Science, Okayama University, Okayama 700-8530, Japan;

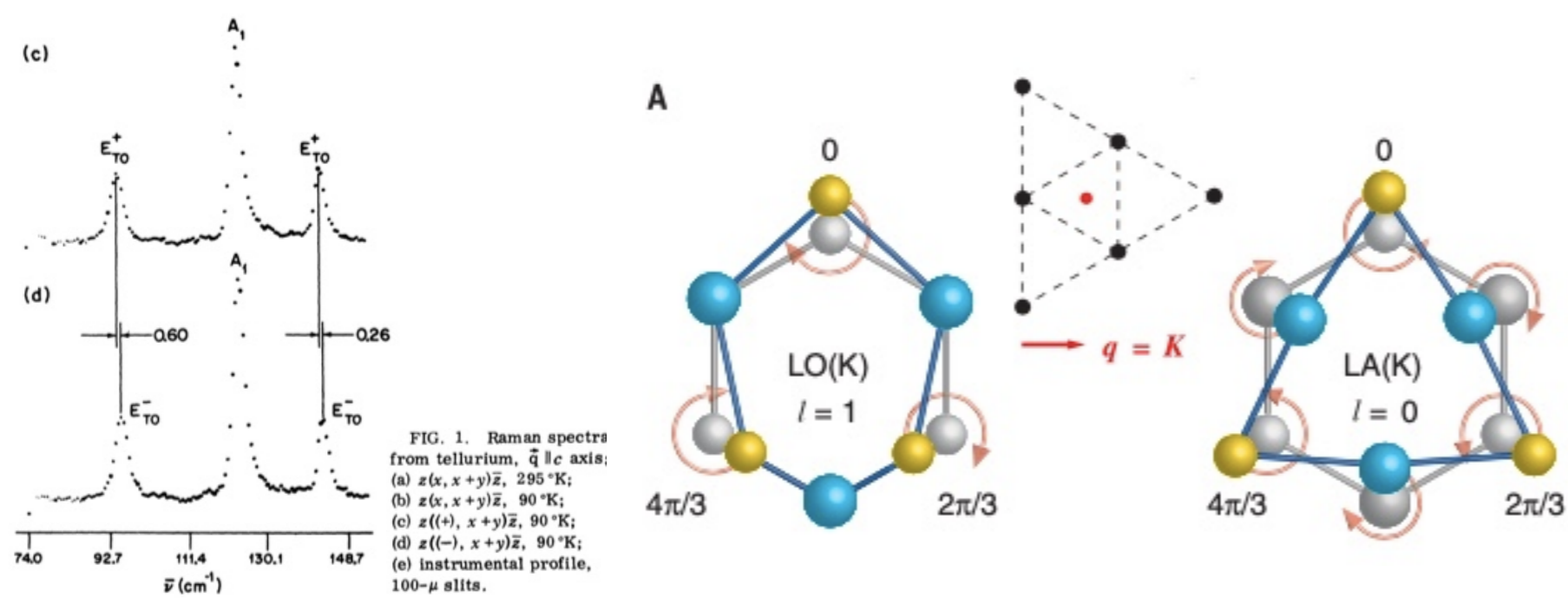
³The Open University of Japan, Chiba, 261-8586, Japan)

Introduction

Objective

• Determination of handedness of helical single crystal Te by chiral phonon.

Background



E-mode phonon splitting in α -quartz

A. S. Pine et al. Phys. Rev. **188**, 1489 (1969).

Chiral phonon in WSe₂

H. Zhu et al. Science **359**, 579 (2018).

Chiral phonon

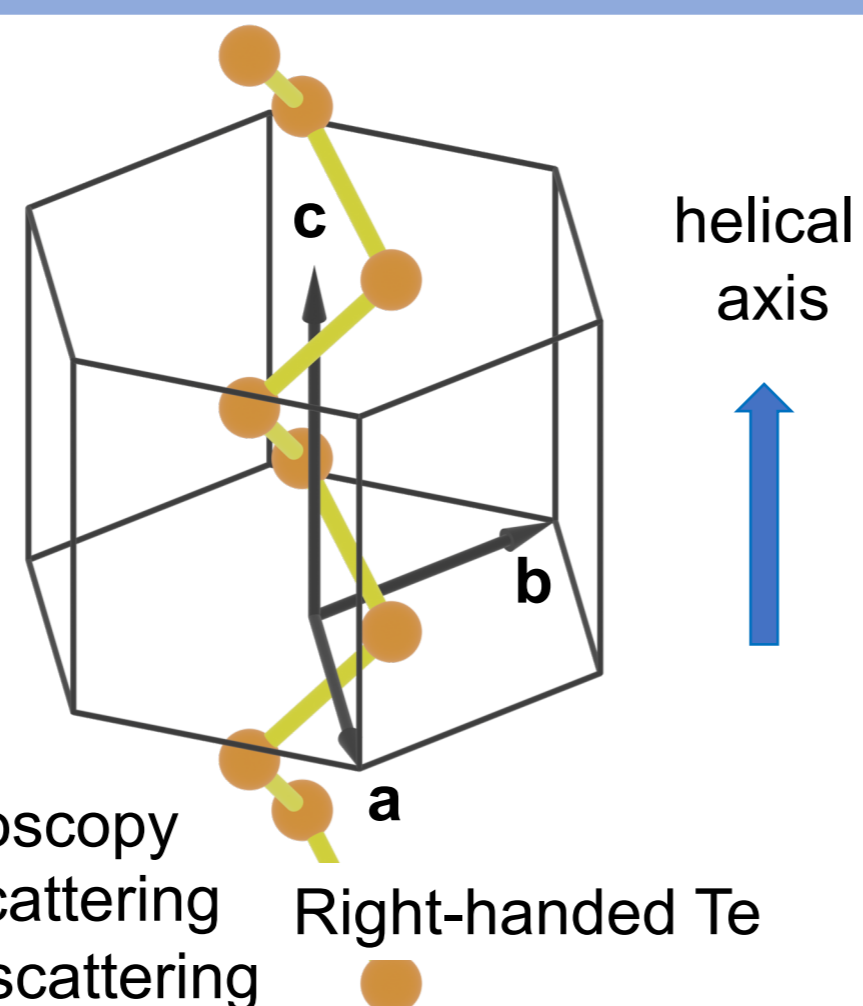
Phonons which have finite angular momentum and pseudoangular momentum (PAM)

Material and Method

Material

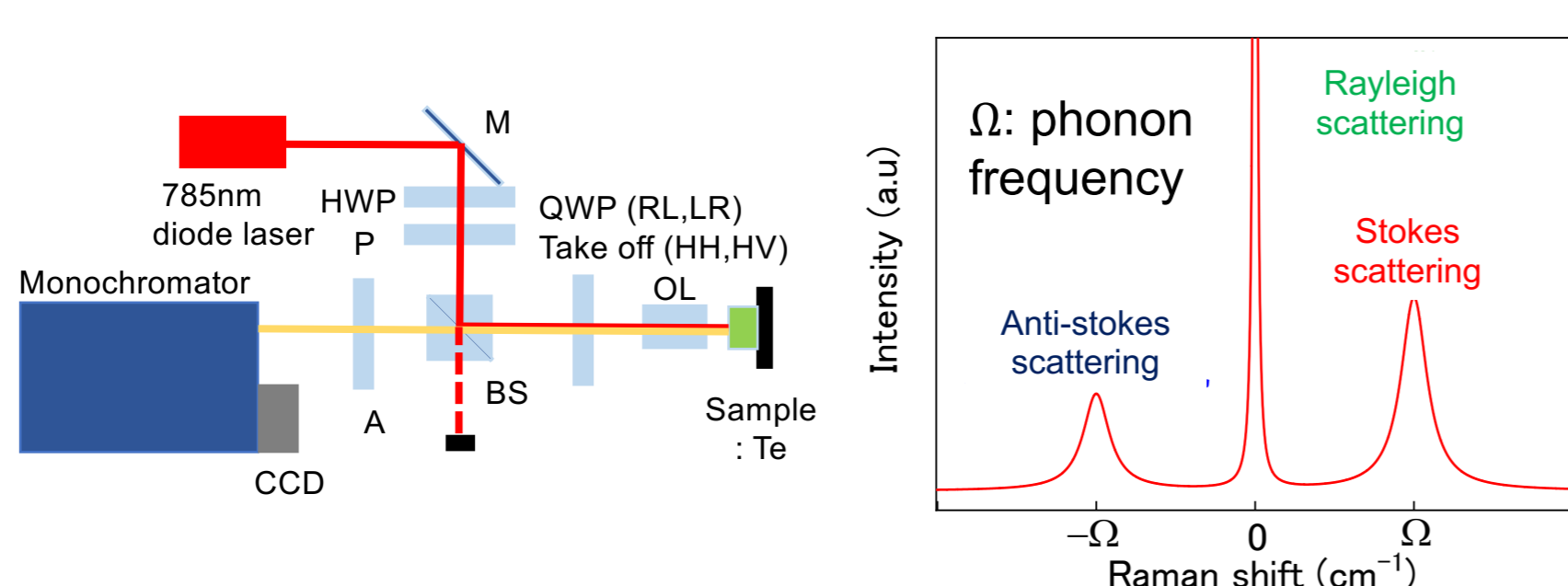
Te (Tellurium)

- lack of inversion symmetry and mirror symmetry
- two enantiomeric structures (P3₁21, Right-handed and P3₂21, Left-handed)
- helical axis
- (3₁, Right-handed, 3₂, left-handed)



Method

1. Circularly polarized Raman spectroscopy
 - Inelastic circularly polarized light scattering
 - photon frequency change through scattering corresponds phonon frequency



Experimental schematic

Raman spectrum

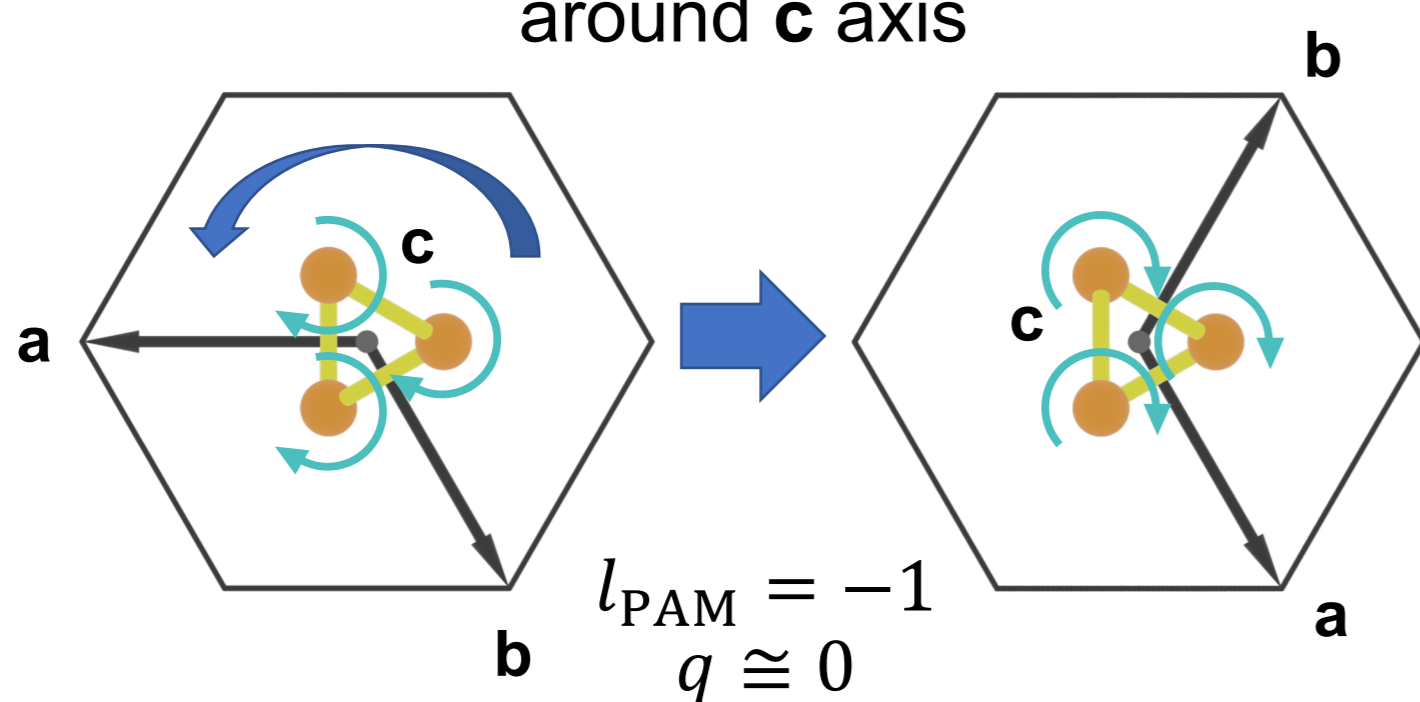
2. PAM calculation

• PAM originates from phase change by discrete helical rotational symmetry operation.

$$\left[C_3 \left| \frac{1}{3} \right. \right] u = \exp \left[-i \left(\frac{2\pi}{3} l_{\text{PAM}} + \frac{q \cdot c}{3} \right) \right] u$$

$\left[C_3 \left| \frac{1}{3} \right. \right]$: three fold helical rotation, l_{PAM} : pseudoangular momentum, q : wave number of phonon, u : eigenvector of dynamical matrix

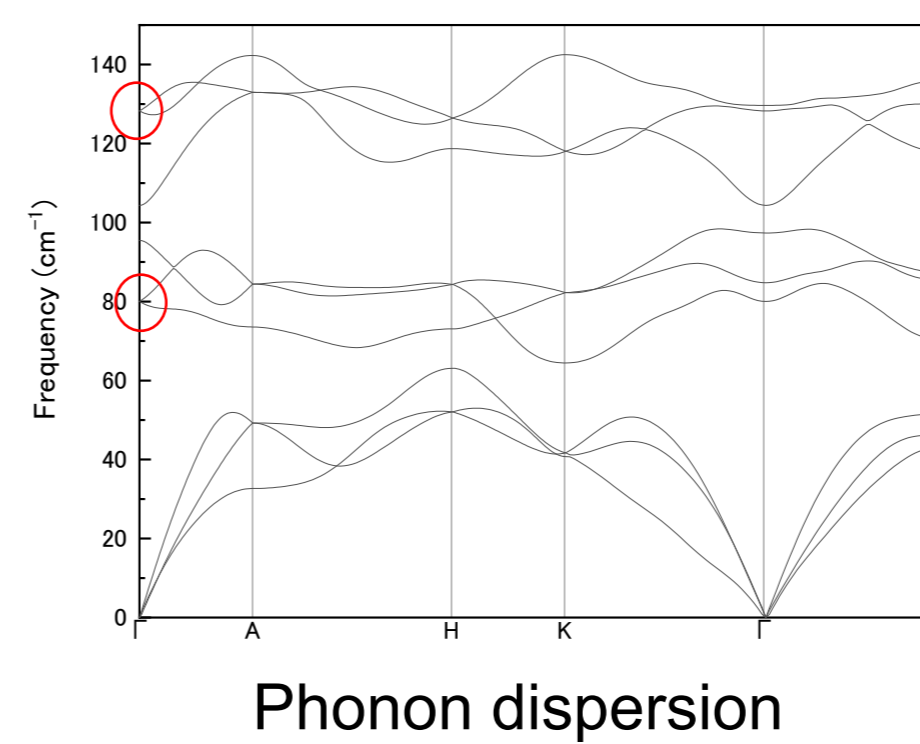
Three fold helical rotation around **c** axis



Result and Discussion

Result

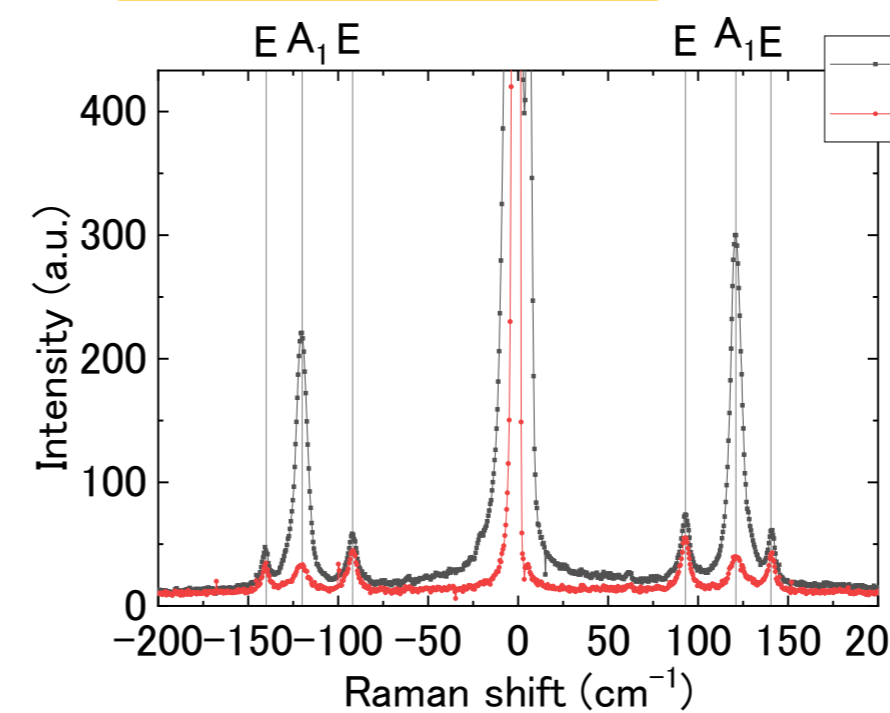
Calculation



Phonon dispersion

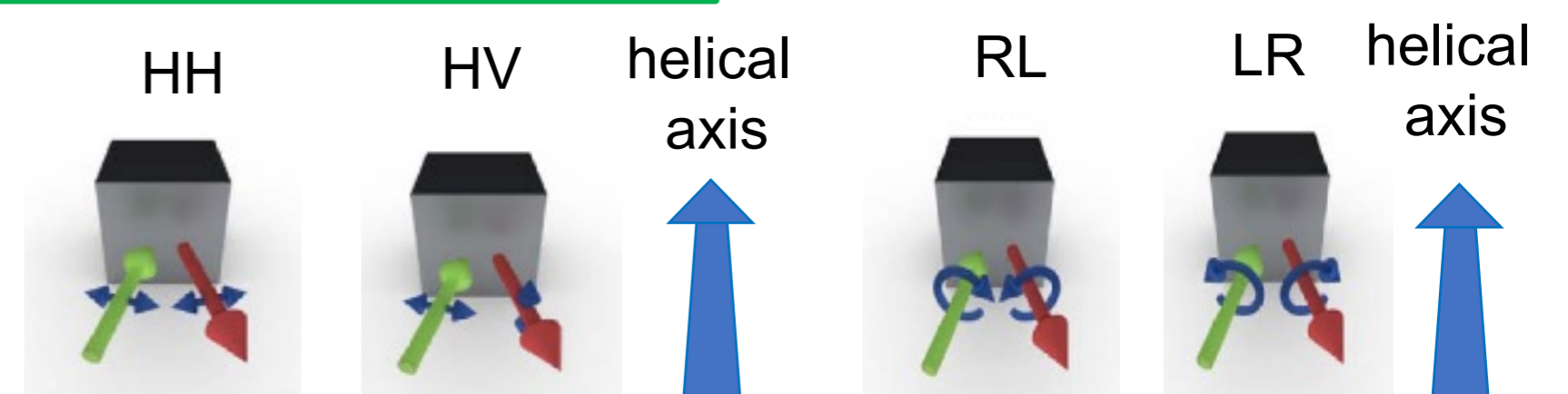
Right-handed Te

Experiment



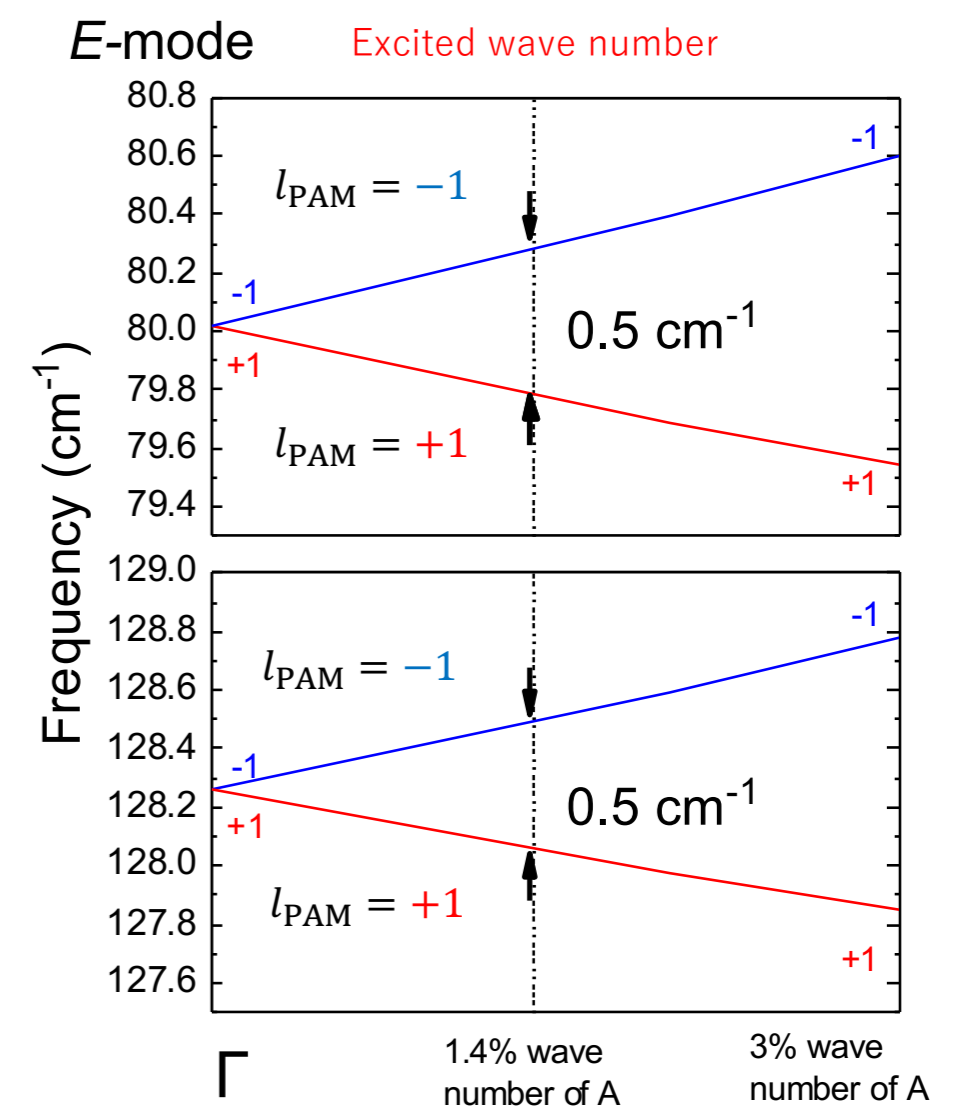
Raman spectrum

unknown handedness



Back scattering along c axis with linear and circular polarization

W. -H. Hsu et al. Phys. Rev. B **102**, 174432 (2020).



- A₁ and 2E modes are observed.
- E-modes split in RL and LR configuration.
- phonons at 1.4% wavenumber of A point are observed by momentum conservation law between photons and phonon.

Discussion

1. PAM conservation law :

$$\sigma_i - \sigma_s \equiv l_{\text{PAM}} \pmod{3}$$

$\sigma_{i,s}$: PAM of incident and scattering photons (R : +1, L : -1)

$$\text{RL} : l_{\text{PAM}} \equiv +1 - (-1) \equiv -1$$

$$\text{LR} : l_{\text{PAM}} \equiv -1 - (+1) \equiv +1$$

2. PAM of right-handed Te and experiment of unknown handedness Te are consistent.

PAM of left-handed Te is opposite in sign to that of right-handed Te.

→ We observe right-handed Te.

3. New method of chirality determination

- Non-destructive
- No solution needed.

Conclusion

Conclusion

- We measured chiral phonons in helical single crystal Te by circularly polarized Raman spectroscopy along the helical axis.
- E-mode split into two modes which are observed in RL and LR configuration, respectively.
- We identified handedness of helical single crystal Te by pseudoangular conservation law in Raman scattering process by comparison between experiment and *ab initio* calculation.