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Anomalous mode, elastic constants and phonon images in CaWO_4 ¹K. Hayasaka, ²R. Higashi, ³J. Suda, ¹Y. Tanaka, ¹S. Tamura, ⁴M. Giltrow and ⁴J. K. Wigmore¹*Department of Applied Physics, Hokkaido University, Sapporo 060-8628,**Japan,* ²*Engineering Research Institute, The University of Tokyo,**Tokyo 113-8656, Japan,* ³*Kushiro National College of Technology,**Kushiro 060-8628, Japan,* ⁴*Department of Physics,**University of Lancaster, Lancaster LA14YB, United Kingdom*

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Stimulated by the “anomalous mode” observed in the time-of-flight measurement of phonon signals [J. K. Wigmore *et al.*, Physica B **316-317**, 589 (2002)], we have theoretically studied the group velocities and the phonon images in CaWO_4 . Comparing the phonon caustics of transverse modes in the low-frequency nondispersive and slightly dispersive 1-THz regions, we propose that the anomalous phonon mode is originated from the phonons near the cuspidal edge of the group velocity curve of the fast transverse branch. Also the low-frequency phonon images (two-dimensional maps of phonon group velocities) calculated from the two sets of published elastic-constant data of CaWO_4 [Farley *et al.*, J. Phys. C: Solid State Phys. **5**, 3021 (1972), and Gluyas *et al.*, J. Phys. D: Appl. Phys. **6**, 2025 (1973)] are compared. Significant difference is found in the images in the X-Y plane due to the variance of the reported values of C_{13} of about 40 percent.

Calcium tungstate (CaWO_4) has a scheelite structure of a tetragonal crystal symmetry belonging to the Laue group TII (crystal class $4/m$) and is an important material in laser technology and also for acousto-optic applications. It is currently utilized in CRESST (cryogenic rare event search with superconducting thermometers) experiment for a dark matter search, where CaWO_4 crystals act as “target” crystals for simultaneous measurement of scintillation light and phonons.[1] Thus the precise information on the acoustic phonon properties from the low-frequency nondispersive sub-THz to slightly dispersive 1-THz regions should be important.

Recently, Wigmore *et al.* made time-of-flight measurements of thermally generated nonequilibrium phonons in CaWO_4 and they found the existence of an “anomalous” signal that cannot be explained by the acoustic measurements based on the sound velocities.[2] The propagation of pulsed phonons (a heat pulse) is governed by the group velocities of phonons rather than the sound velocities (phase velocities). So, the “anomalous” signal reported must be an effect related to the phonon group velocities which are highly noncollinear with a given wave vector direction, in general, affected strongly by the elastic anisotropy of the crystal. Thus in the present work we try to explore the origin of the anomalous phonon mode (reported in ref. 2) based on the analysis of the group velocities of three phonon polarizations in CaWO_4 . Explicitly, this will be done by calculating low-frequency phonon group velocities based on the published elastic constants and also by studying the frequency dependence of the group-velocity curves up to about 1 THz with a lattice dynamics model.

Here we briefly recapitulate the heat pulse experiment of Wigmore *et al.* They measured the transmission of phonons across the shortest dimension of an $18 \text{ mm} \times 6 \text{ mm} \times 3 \text{ mm}$ CaWO_4 sample.[2] The corresponding propagation direction (we call the b axis) is not along

the crystallographic axis. The sample axes are rotated from the crystallographic X , Y and Z axes as shown in the inset of Fig. 1. First, the X and Y axes are rotated about the four-fold Z axis by $\alpha = 7 \pm 1^\circ$ and then Z and new Y axes are rotated about the new X axis (a axis) by $\beta = 5 \pm 1^\circ$. We call the resulting axes originated from the X , Y and Z axes the a , b and c axes, respectively.

To analyze the time-of-flight data, first, we have used the elasticity model and calculated for a given wave vector $\mathbf{k}$ both the phase velocity $c = \omega/|\mathbf{k}|$ (with ω the angular frequency) and group velocity $\mathbf{v} = \partial\omega/\partial\mathbf{k}$ for three phonon modes by solving the Christoffel equations

$$(\rho\omega^2\delta_{im} - C_{ilmn}k_l k_n)e_m = 0, \quad (i = 1, 2, 3), \quad (1)$$

where ρ is the mass density, C_{ilmn} is the stiffness tensor and $\mathbf{e}$ is the polarization vector (the summation convention over repeated indices is assumed).

Figure 1 shows the group velocity curves of three phonon modes in the $a - b$ plane calculated from two sets of published elastic constant data for CaWO_4 . Here we note that the experimental determination of seven independent elastic constants in CaWO_4 has been done independently by Farley *et al.*[3, 4] and by Gluyas *et al.*[5] with ultrasound-velocity measurements over a wide range of temperatures 1-300 K. Although these two sets of elastic constants are quite similar as we expect (see the Table I), there exist some quantitative differences in C_{13} and C_{66} , up to about 40 % and 20 %, respectively.

For the propagation along the b axis there exist three phonon branches and the outermost longitudinal (L) and two inner transverse (T) branches are found at the velocities of $5.01(5.01) \times 10^5 \text{ cm/s}$, $2.42(2.42) \times 10^5 \text{ cm/s}$ and $1.94(2.22) \times 10^5 \text{ cm/s}$, respectively, with the elastic-constant data by Farley (Gluyas) *et al.* Thus, the overall coincidence of the calculated group velocities is very well,

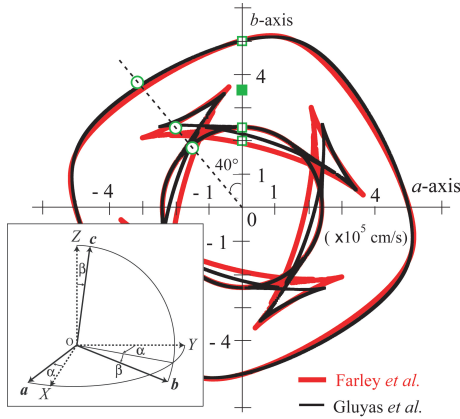


FIG. 1: (color on line) Group velocity curves in the $a - b$ plane of CaWO_4 calculated with the low-temperature elastic constants given by Farley *et al.* (bold lines) and Gluyas *et al.* (thin lines). The positions of the intensity peaks observed in the time-of-flight spectra along the b axis and those along the 40° direction rotated from the b axis are converted to velocities and shown by the open squares and circles, respectively. The point labeled by the filled square indicates the position of the anomalous peak. Inset shows the definition of the sample axes (a , b and c axes) to the crystallographic X , Y and Z axes with $\alpha = 7 \pm 1^\circ$ and $\beta = 5 \pm 1^\circ$.

though the cuspidal structures of the T mode obtained from the data of Farley *et al.* are a bit larger and there exist small deviations in the innermost branches continuing from these cusps.

The phonon velocities deduced from the experimentally observed peaks in the time-of-flight spectra along the b axis [2] and also in the direction 40° rotated from b axis in the $a - b$ plane (“ 40° experiment” [6]) are shown in Fig. 1 by the open squares and circles, respectively. Their positions are well overlapped with the theoretical group velocity curves except for the one labeled with a filled square located between the L and T branches. Wigmore *et al.* called the signal found at this point “anomalous mode”. We see that the corresponding position is close to a cuspidal edge of the low-frequency T phonons (more precisely the FT phonons [7]) predicted from the elastic-constant data.

An interesting experimental result reported is the fact that the anomalous mode peak depends sensitively on the excitation power of the heat pulse.[2] At the lowest power levels, where we believe that low-frequency, nondispersive phonons are predominantly excited, this peak is not seen at all. Accordingly, this is consistent with our calculated results (Fig. 1) of the phonon group velocities in the continuum approximations. As heater power increases very rapid increase of the signal is observed.[8] Thus our expectation is that this power dependence of the anomalous mode should be the effect related to the lattice dispersion, that is, to the changes of phonon group velocities with increasing frequency. More explicitly the cusps in the FT phonon group velocities near the b axis may ex-

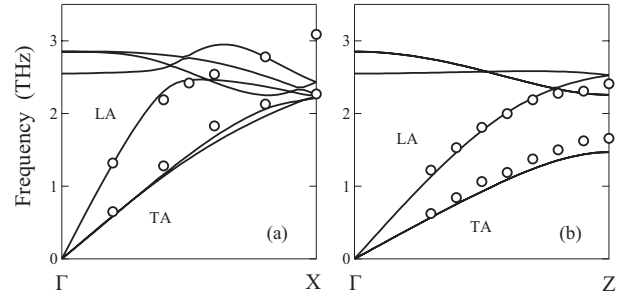


FIG. 2: Phonon dispersion relations (below about 3 THz) in CaWO_4 , (a) in the Γ -X direction and (b) in the Γ -Z direction. The experimental (circles) [12] and calculated (solid lines) frequencies for the transverse acoustic (TA) and longitudinal acoustic (LA) branches are compared.

tend and crosses this axis as phonon frequency increases. In order to see if this happens or not, next we try to do lattice dynamics calculations.

The lattice dynamics for CaWO_4 have been developed by several groups to calculate phonon dispersion curves and also to study the thermal properties. [9–11] Here we employ the recently published model by Suda and Sato [10] based on the rigid-ion model. As shown in Fig. 2, the coincidence between the predicted and experimental acoustic phonon frequencies is qualitatively acceptable in the Γ -X and Γ -Z directions below about 2 THz. [12]

In Fig. 3 we have displayed how the group velocity curve of the FT phonons in the upper $a - b$ plane changes with frequency.[13] We see that the FT focusing cusp expands and moves toward the b axis as frequency increases (for $\nu > 0.5$ THz) but does not cross this axis possibly because the magnitude of the calculated FT cusps has not been reproduced very quantitatively at the low-frequency limit. Taking account of the uncertainty in the estimation of the crystal directions and slight inaccuracy in the lattice dynamics model for the prediction of low-frequency acoustic branches, it should be reasonable to conclude that the rapid increase observed in the detected anomalous signal with heater power is the effect arising from the frequency-shifted cusp of the FT phonons.

So far we have neglected the effects of phonon scattering which become important as frequency increases. The elastic scattering of phonons (possibly due to the isotopic atoms in CaWO_4) is known to extend the focusing ridges of the phonon intensity outside the caustics.[14] Hence, the elevation of the phonon signal as seen in the power dependence of the anomalous peak may happen before the edge of the FT cusp touches the b axis. So, we have also made a preliminary Monte Carlo calculation for the phonon propagation with both the elastic anisotropy and scatterings due to isotopic atoms in CaWO_4 . [15–17] The scattering rate used for the simulation is $\tau_I^{-1} = 5.90 \times 10^{-41} \nu^4 \text{ s}^{-1}$. [18] However, no sharp rise of phonon signal has been found at the expected arrival time of the anomalous mode, though some moderate increase of the intensity is seen. The situation has not

changed qualitatively even if we further included spontaneous phonon decays (due to three-phonon processes in the isotropic approximation).[19] Thus the phonon scatterings could have only secondary effect as far as the origin of the anomalous signal is concerned.

Finally, how can we reconcile the variance of two elastic constants C_{13} and C_{66} obtained experimentally by two groups? As we have seen in Fig. 1 the phonon group velocities calculated with the data of Farley *et al.* and Gluyas *et al.* have some quantitative difference (e. g., the magnitude of the FT cusps) mainly due to the measured variance of C_{13} and C_{66} . However, it may be rather hard to discriminate the magnitudes of those group velocities by a spectroscopic experiment such as a time-of-flight measurement. Here we note that there exists another powerful method to characterize the phonon group velocities through their angular dependences. This is the phonon imaging experiment.[14] The phonon imaging has primarily been used to reveal strong focusing effects of ballistic phonons arising from the elastic anisotropy of crystalline solids. [20, 21] For a number of crystals phonon images have so far been obtained with various techniques and the coincidence between the experimental and theoretical images (calculated with accepted elastic constant data) are excellent, in general, as far as the low-frequency, nondispersive phonons are concerned.[14]

Figures. 4(a) and 4(b) show the phonon images calculated with two sets of elastic constants by Farley *et al.* and Gluyas *et al.*, and they demonstrate even more dramatic difference (than the one in Fig. 1) in the group

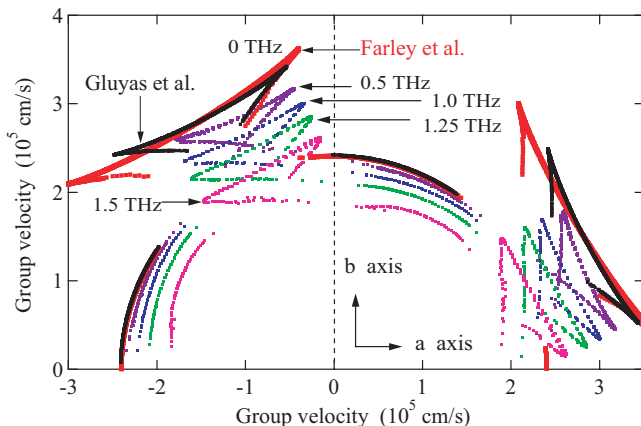


FIG. 3: (color on line) Comparison of the group velocity curves in the upper $a - b$ plane of the fast transverse (FT) phonons calculated with the elastic constants measured by Farley *et al.* and Gluyas *et al.* (0 THz), and with the lattice dynamics model for 0.5 THz, 1.0 THz, 1.25 THz and 1.5 THz frequencies. The calculated dispersion curves of the transverse acoustic (TA) branches are located a bit lower than the experimental frequencies (Fig. 2), leading to the group velocities smaller than those obtained from the elasticity theory. The calculated group velocities are rescaled by multiplying a factor of 1.1 so that the magnitude of the TA phonon group velocities at 0.5 THz may become comparable to the ones in Fig. 1.

velocity distribution of phonons in the four-fold $X - Y$ plane of CaWO_4 . The difference has been emphasized because phonon group velocities and the subsequent ballistic phonon images are determined from the curvatures of the constant-frequency surfaces of three phonon polarizations and hence they are very sensitive to the measured values of the elastic constants in the low-frequency limit. At a glance, we recognize marked contrast in both the shapes and widths of the inner and outer focused regions of phonons surrounded by the caustics.[22] We have also made similar calculations for the images in the $X - Z$ plane and found large contrast as well. By changing artificially the magnitudes of several elastic constants we have confirmed that the origin of these contrasts in the calculated phonon images are attributed to the variance of C_{13} . The phonon images in the both planes are rather insensitive to the variation of C_{66} .

To summarize, we have calculated the phonon group velocities in CaWO_4 with published data of elastic constants to locate the origin of the anomalous phonon signal reported by Wigmore *et al.* We have presented a possible evidence that the signal can be attributed to the FT phonons which have a large cusp near the b -axis in the group velocity curves. As the phonon frequency increases, the calculated cuspidal edge of FT phonons is found to get closer to the b axis along which the anomalous peak was observed at high excitation power levels. Quantitative details of the measured power dependence of the anomalous signal could be obtained by theoretical analysis with a more elaborate lattice dynamics model taking into account the sizes of heater and bolometer, [23] and the effects of scattering. However, only a direct phonon imaging experiment could give details necessary for a full specification of the model and elastic constants. Such an experiment is urgently needed in order to characterize fully the propagation of ballistic and scattered high-frequency acoustic phonons in CaWO_4 important for the dark matter search.

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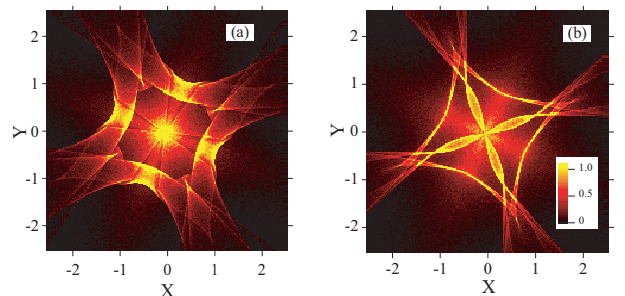


FIG. 4: (color on line) Phonon images in the X - Y plane of CaWO_4 calculated with the elastic constants of (a) Farley *et al.* and (b) Gluyas *et al.* The brightness corresponds to the concentration of phonon group velocities. The horizontal and vertical axes measure the positions in the $X - Y$ plane when the phonons are emitted ballistically from the point source by a unit distance below the origin of this plane.

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TABLE I: The elastic constants in units of GPa measured at low temperatures (indicated in the parentheses). The mass density of CaWO₄ is $\rho = 6.12 \text{ g cm}^{-3}$. The sign of C_{16} not determined by Gluyas *et al.*[5] is assigned so that the calculated shapes of the slowness and group velocity curves of phonons are consistent with those obtained by Farley *et al.*[3]

	C_{11}	C_{12}	C_{13}	C_{16}	C_{33}	C_{44}	C_{66}
Farley <i>et al.</i> (1.5 K)	152.3	65.2	40.8	-20.4	132.4	35.2	40.3
Gluyas <i>et al.</i> (4.2 K)	150.6	65.9	58.9	-17.2	135.6	35.6	47.7