

Lattice dynamics and complete polarization analysis of Raman-active modes in LaInO_3

Jonas Rose^{1,*}, Hai Nguyen¹, Moritz Meißner¹, Zbigniew Galazka², Roland Gillen³, Georg Hoffmann¹, Oliver Brandt¹, Manfred Ramsteiner¹, Markus R. Wagner¹, and Hans Tornatzky^{1,†}

¹*Paul-Drude-Institut für Festkörperelektronik, Leibniz-Institut im Forschungsverbund Berlin e. V., Hausvogteiplatz 5–7, 10117 Berlin, Germany*

²*Leibniz-Institut für Kristallzüchtung, Max-Born-Strasse 2, 12489 Berlin, Germany*

³*Department of Electronic and Electrical Engineering, Faculty of Science and Engineering, Swansea University, Swansea SA1 8EN, Wales, United Kingdom*



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In this study, we present a comprehensive analysis of the Raman-active phonon modes in orthorhombic LaInO_3 based on a combination of polarization-angle resolved Raman spectroscopy and density functional theory calculations. By using backscattering from multiple crystallographic surface orientations and employing a full symmetry analysis, we identify and assign most of the Raman-active Γ -point phonons to their irreducible representations of the D_{2h} point group. A multidimensional hyperspectral fitting procedure allows us to extract the relative Raman tensor elements from the angular dependence of the scattering intensities, even for strongly overlapping modes. First-principles calculations yield the phonon dispersion along high-symmetry directions, the phonon densities of states, atomic displacement patterns, and Raman tensor elements, which are found to be in good agreement with the experiment.

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I. INTRODUCTION

Over the last years, transparent conductive oxides have become a fundamental building block for new device concepts and an important topic of fundamental research. In particular, perovskite oxides (ABO_3) exhibit multifunctional physical properties, spanning superconductivity, magnetism, and topological conductivity among others. Even more exciting, their ability to epitaxially stack on top of each other generates new physical phenomena at their interfaces beyond their bulk properties, like two-dimensional (2D) superconductivity [1], 2D magnetism [2], or 2D electron gases (2DEGs) [3].

Within this family, La-doped BaSnO_3 (BSO) has emerged as a key material. Single crystal BSO exhibits the highest room-temperature electron mobility ($\mu \approx 320 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ [4]) reported for perovskite oxides while maintaining excellent optical transparency due to its wide band gap of 3.1 eV. It further provides exceptional thermal stability [5] and a low oxygen diffusion coefficient [6]. In epitaxial thin films, however, the mobility is typically reduced when using commercially available substrates such as SrTiO_3 [7], LaAlO_3 [7], MgO [8], or DyScO_3 [9,10]. This reduction is widely attributed to scattering at threading dislocations induced by substantial lattice mismatch of up to 5% as well as

crystallographic shear defects and point defects arising from nonstoichiometry [9,11]. Beyond advances in epitaxial growth, employing substrates with improved lattice compatibility therefore represents a promising strategy to enhance the electronic performance of BSO heterostructures.

To this end, LaInO_3 (LIO) has emerged as one of the most promising materials as it exhibits a pseudocubic lattice parameter that is nearly matched to that of BSO and can be grown from the melt [12]. More interestingly, it was demonstrated that a $(\text{SnO}_2)^0/(\text{LaO})^{+1}$ -terminated BSO/LIO interface gives rise to a 2DEG confined in the BSO through polar discontinuity doping [7,13]. Moreover, by employing LIO as a gate oxide, field effect transistors can be realized that offer high electron mobility, low sheet resistance, and high charge carrier densities, positioning the BSO/LIO heterostructure as a compelling platform towards novel oxide electronic devices.

Despite the increasing relevance of LIO in this context, detailed investigations of its lattice dynamics remain scarce as previous studies suffered from inferior sample quality [14,15]. As phonon-related mechanisms are one of the most important foundations for understanding materials, covering mechanical and elastic properties, thermal transport, as well as charge-carrier dynamics and phonon-assisted optical excitations, a comprehensive understanding of the lattice dynamics is essential for the proper design of associated processes in any application.

In this study, we investigate the lattice dynamics of bulk LIO by combining polarization-angle resolved Raman spectroscopy with density functional theory (DFT). Experimentally, we observe 19 of the 24 Raman-active modes and assign them to irreducible representations of the crystal's point group based on a symmetry analysis. By employing a multidimensional fitting procedure, we determine the relative

*Contact author: rose@pdi-berlin.de

†Contact author: tornatzky@pdi-berlin.de

TABLE I. Wyckoff positions with site symmetry [16] and fractional coordinates of the atoms, calculated from DFT, and their contributions to the irreducible representations of the Γ -point phonons in the $Pnma$ space group [17].

Atom	Site	Symmetry	Fractional coordinates			Raman-active				IR-active and acoustic			Silent
			x	y	z	A_g	B_{1g}	B_{2g}	B_{3g}	B_{1u}	B_{2u}	B_{3u}	
La	4c	C_s	0.9442	0.2500	0.0170	2	1	2	1	2	1	2	1
In	4b	C_i	0.0000	0.0000	0.5000	—	—	—	—	3	3	3	3
O(1)	4c	C_s	0.0498	0.2500	0.6186	2	1	2	1	2	1	2	1
O(2)	8d	C_1	0.1970	0.0628	0.1947	3	3	3	3	3	3	3	3

Raman tensor elements of the observed modes from the angular dependencies of their corresponding measured scattering efficiencies. Furthermore, we present the DFT-derived phonon dispersion along high-symmetry paths in reciprocal space, visualize the atomic displacement patterns for all Γ -point phonons, and compare the calculated mode frequencies with our experimental observations.

II. CRYSTAL STRUCTURE AND RAMAN-ACTIVE PHONONS

LIO crystallizes in an orthorhombic crystal structure and belongs to the space group D_{2h}^{16} in Schönflies notation (no. 62 in International Tables for Crystallography, ITC). There are two common Hermann-Mauguin notations for this space group, $Pnma$ and $Pbnm$, which differ only by a cyclic permutation of the lattice vectors. To remain consistent with the ITC, all notations in this study follow the $Pnma$ setting, in which the lattice constant b_0 (along the B axis) is the longest. The crystal is fully described by a primitive orthorhombic Bravais lattice with lattice parameters of $a_0 = 5.938$ Å, $b_0 = 8.214$ Å, and $c_0 = 5.723$ Å [12] and a basis which is constructed using the Wyckoff sites and fractional coordinates given in Table I.

The atomic basis comprises 20 atoms ($Z = 4$), giving rise to 60 phonon modes. At the Γ point, the corresponding atomic displacements decompose into the irreducible representations of the D_{2h} point group [17]:

$$\Gamma = 7A_g \oplus 5B_{1g} \oplus 7B_{2g} \oplus 5B_{3g} \\ \oplus 10B_{1u} \oplus 8B_{2u} \oplus 10B_{3u} \oplus 8A_u.$$

Of these modes, 24 ($7A_g \oplus 5B_{1g} \oplus 7B_{2g} \oplus 5B_{3g}$) are Raman active, 25 ($9B_{1u} \oplus 7B_{2u} \oplus 9B_{3u}$) are IR active, three ($B_{1u} \oplus B_{2u} \oplus B_{3u}$) are acoustic, and eight ($8A_u$) are silent. In the following, Raman-active modes are labeled with a superscript index that reflects their energetic ordering within each irreducible representation. As summarized in Table I, In atoms do not contribute to Raman-active modes as their site symmetry includes an inversion center, allowing participation only in odd-parity modes for the LIO crystal structure.

Table II summarizes the selection rules for all Raman-active phonon modes in the four different measurement geometries available to us. The Raman tensors R are expressed in the crystal principal axis system (A, B, C), which is aligned with the fixed laboratory coordinate system (X, Y, Z). If a different axis is oriented along Z , i.e. if another surface plane is investigated, the transformed Raman tensors are obtained by applying the appropriate rotation matrices. In our experiments, the Z axis is parallel to both the incident and scattered

light, while the polarization is rotated within the XY plane by an angle ϕ . The corresponding polarization vectors are given by $\hat{e}_i = (\cos(\phi), \sin(\phi), 0)$ and $\hat{e}_s = (\cos(\phi), \sin(\phi), 0)$ for parallel polarization or $\hat{e}_s = (-\sin(\phi), \cos(\phi), 0)$ for crossed polarization. Under these conditions, the third row and column of the transformed Raman tensors as well as the third entries of $\hat{e}_{i,s}$ can be eliminated and the problem becomes two-dimensional.

Usually, the Raman intensity can be expressed as

$$I \propto |\hat{e}_s R \hat{e}_i|^2. \quad (1)$$

Due to birefringence, i.e. different refractive indices along different axes, the portion of the sample above the scattering event can be considered as a waveplate, which leads to an additional depth-dependent rotation of $\hat{e}_i$ and $\hat{e}_s$. However, for excitations within the transparency regime of bulk crystals, where sufficiently large penetration depths are reached, and considerable birefringence, the depth dependency can be neglected and the Raman intensity is calculated as [18]

$$I \propto |\hat{e}_s R_0 \hat{e}_i|^2 + |\hat{e}_s R_1 \hat{e}_i|^2 + |\hat{e}_s R_2 \hat{e}_i|^2, \quad (2)$$

where

$$R_0 = \begin{pmatrix} r_0 & 0 \\ 0 & 0 \end{pmatrix}, \quad R_1 = \begin{pmatrix} 0 & r_1 \\ r_1 & 0 \end{pmatrix}, \quad R_2 = \begin{pmatrix} 0 & 0 \\ 0 & r_2 \end{pmatrix} \quad (3)$$

are summands of the transformed Raman tensor, such that $R = R_0 + R_1 + R_2$.

Note that birefringence has no influence on the B_{xg} modes in the geometries listed in Table II. As their transformed Raman tensors only possess off-diagonal elements, R_0 and R_2 vanish and the expression in Eq. (2) collapses to a single term identical to Eq. (1). Furthermore, birefringence affects the transmission of light at the air-sample interface, resulting in an additional angle-dependent modulation of the scattering intensity. This effect is taken into account by multiplying the transformed Raman tensor from left and right by the diagonal matrix $\text{diag}(t_x/t_y, 1)$ [19], effectively scaling r_0 and r_1 from Eq. (3) by $(t_x/t_y)^2$ and t_x/t_y , respectively. Here, $t_e = 2/(n_e + 1)$ denotes the Fresnel transmission coefficient for light propagating along Z and polarization along $\hat{e}$.

In order to compare the tensor elements of different modes with each other as well as with theoretical calculations, a wave-number-dependent prefactor [20]

$$F(\omega_p) = \frac{\omega_L(\omega_L - \omega_p)^3}{\omega_p [1 - \exp(-\frac{\hbar\omega_p}{k_B T})]} \quad (4)$$

for the measured scattering intensity must be considered, where ω_L is the laser and ω_p is the phonon angular frequency.

TABLE II. Selection rules of all Raman-active phonon modes in four different geometries including plots of their intensity profiles. The first row of each geometry corresponds to parallel polarization (blue), and the second row corresponds to crossed polarization (orange). The Raman tensors R are expressed in the crystal principal axis (A, B, C). N denotes the surface normal of the (101) face. The ratios of the Raman tensor elements a, b , and c in the plotted A_g mode intensity profiles are 10 : 7 : 8.

Geometry	A_g $\begin{pmatrix} a & 0 & 0 \\ 0 & b & 0 \\ 0 & 0 & c \end{pmatrix}$		B_{1g} $\begin{pmatrix} 0 & d & 0 \\ d & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$		B_{2g} $\begin{pmatrix} 0 & 0 & e \\ 0 & 0 & 0 \\ e & 0 & 0 \end{pmatrix}$		B_{3g} $\begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & f \\ 0 & f & 0 \end{pmatrix}$	
$A \parallel Z$ (100)	$b^2 \sin^4(\phi) + c^2 \cos^4(\phi)$ $\frac{1}{4}(b^2 + c^2) \sin^2(2\phi)$	 	0 0		0 0		$f^2 \sin^2(2\phi)$ $f^2 \cos^2(2\phi)$	 
$B \parallel Z$ (010)	$c^2 \sin^4(\phi) + a^2 \cos^4(\phi)$ $\frac{1}{4}(a^2 + c^2) \sin^2(2\phi)$	 	0 0		$e^2 \sin^2(2\phi)$ $e^2 \cos^2(2\phi)$	 	0 0	
$C \parallel Z$ (001)	$a^2 \sin^4(\phi) + b^2 \cos^4(\phi)$ $\frac{1}{4}(a^2 + b^2) \sin^2(2\phi)$	 	$d^2 \sin^2(2\phi)$ $d^2 \cos^2(2\phi)$	 	0 0		0 0	
$N \parallel Z$ (101)	$b^2 \cos^4(\phi) + (a' + c')^2 \sin^4(\phi)$ $\frac{1}{4}(b^2 + (a' + c')^2) \sin^2(2\phi)$	 	$d'^2 \sin^2(2\phi)$ $d'^2 \cos^2(2\phi)$	 	$e^2 \sin^4(\phi)$ $\frac{1}{4}e^2 \sin^2(2\phi)$	 	$f'^2 \sin^2(2\phi)$ $f'^2 \cos^2(2\phi)$	 

$a' = a \cos^2(\theta), c' = c \sin^2(\theta), d' = d \cos(\theta)$
 $e' = e \sin(2\theta), f' = f \sin(\theta), \theta = \arctan(\frac{c_0}{a_0})$

III. METHODS

A. Raman spectroscopy

Raman measurements were performed with a Horiba LabRAM HR Evolution Raman spectrometer using linearly polarized 632.8-nm HeNe laser excitation in back-scattering geometry. A half-wave plate (HWP) on a motorized rotation stage allows switching of the polarization angle between $\hat{e}_i$ and $\hat{e}_s$ from 0° to 90° , enabling parallel and crossed polarization configurations. After reflection from a notch filter, the beam passes through a second HWP, which gradually rotates the polarization relative to the crystal axes with a step size of 10° . A $10\times$ objective (Olympus, NA = 0.25) focuses the laser onto the sample and collects the back-scattered signal. The low NA was chosen to ensure that the calculated selection rules remain as valid as possible. Passing the scattered light also through the second HWP effectively simulates a rotation of the sample around the axis which is aligned with the Z direction without physically moving it, thereby maintaining the probed spot. A fixed analyzer then selects the desired polarization before the scattered light is led through a 200- μm -wide pinhole and dispersed by a 1800-lines/mm grating offering a spectral resolution of 1.2 cm^{-1} . Finally, the light is detected with a charge-coupled device.

B. Sample characteristics

Samples for the present study were prepared from bulk LaInO_3 single crystals grown from the melt by the vertical gradient freeze method at Leibniz-Institut für Kristallzüchtung [12], cut into wafers with the four main orientations (100), (010), (001), and (101) and polished afterwards. Ellipsometric measurements yield real valued refractive indices of $n_A = 2.0992$, $n_B = 2.0667$, and $n_C = 2.1330$ at an excitation wavelength of 632.8 nm, i.e. there is no absorption and the Raman tensor elements can also be assumed real [21,22].

More details on the growth process and structural quality of the samples can be found in Ref. [12].

In principle, backscattering from the (101) surface allows the observation of all Raman-active phonon modes, whereas pairs of A_g and B_{xg} modes can be detected for the other orientations (see Table II), enabling us to distinguish between modes with similar frequencies but different symmetries. Contrary to LIO grown on a BSO substrate, which shows the formation of differently oriented domains [23], the samples in this study are single-domain crystals, meaning the derived selection rules in Table II apply and no mixed phonon signatures should be expected.

C. DFT calculations

DFT as implemented in the QUANTUM ESPRESSO suite [24] was used with generalized gradient approximation (GGA) using Perdew-Burke-Ernzerhof (PBE) functionals to calculate the phonon dispersion and derive the oscillation patterns of the phonon modes at the Γ point. Pseudopotentials from the Materials Cloud Precision library, SSPP PBE Precision v1.3.0 [25], considering $5d^1 6s^2$ (La), $4d^{10} 5s^2 5p^1$ (In), and $2s^2 2p^4$ (O) as valence configuration, were used with a kinetic cut-off energy of 80 Ry (800 Ry) for the wave function (charge density and potential). Electronic and vibrational properties were calculated on a regular $6 \times 4 \times 6$ k - and q -point grid. The optimized lattice parameters, $a_0 = 6.015 \text{ \AA}$, $b_0 = 8.348 \text{ \AA}$, and $c_0 = 5.776 \text{ \AA}$, are found to be slightly larger than experimental x-ray-diffraction values [12,26], consistent with the known *underbinding* of GGA-PBE-based calculations [27]. The resulting underestimation of bond strengths leads to systematically reduced phonon energies; accordingly, the presented phonon dispersion was uniformly scaled by a factor of 1.023, as determined from comparison with experiment. The phonon density of states (pDOS) was computed on a $50 \times 50 \times 50$ q -point grid.

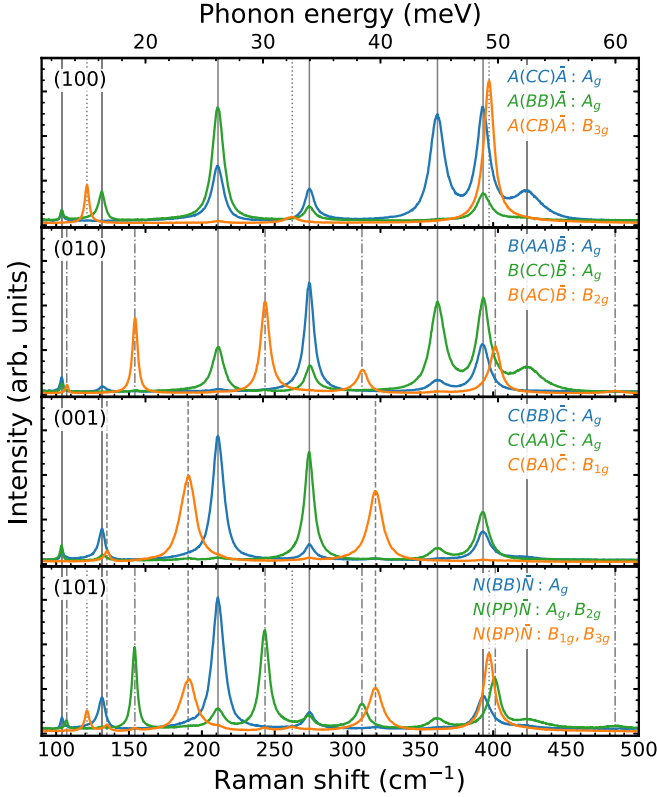


FIG. 1. Polarization-dependent Raman spectra of LIO, recorded with incident and scattered light directed along the A , B , C , and N crystal axes (from top to bottom). Blue and green spectra correspond to parallel polarization configurations, with the polarization vectors rotated by 90° within the XY plane, while orange spectra represent the crossed polarization configuration. The axis P denotes the $[\bar{1}01]$ crystal direction. Vertical solid (A_g), dashed (B_{1g}), dash-dotted (B_{2g}), and dotted (B_{3g}) lines mark the positions of Raman-active phonon modes.

Raman tensor elements were calculated using the code CRYSTAL23 [28], the HSEsol exchange-correlation approximation [29], and pob-TZVP-rev2 basis sets [30]. We optimized the atomic positions until the max rms of the gradient was below 10^{-5} hartree/bohr and the energy change between optimization steps was below 10^{-8} hartree while keeping the lattice constants fixed to the experimental values. To ensure sufficient accuracy of the phonon properties, we used a $6 \times 6 \times 6$ k -point grid and a 12×12 Gilat net, an XXLGRID, and “9 9 9 9 18” as truncation criteria for bielectronic integrals, and a self-consistent field convergence threshold of 10^{-11} hartree for all calculations.

IV. RESULTS AND DISCUSSION

Figure 1 presents the polarized Raman spectra obtained by backscattering from the (100), (010), (001), and (101) crystal faces in three different configurations. They are expressed in Porto’s notation $k_i(e_i e_s)k_s$, where $k_{i,s}$ are the vectors of the incident and scattered light. The polarization vectors $e_{i,s}$ are aligned with relevant crystallographic axes, ensuring that each mode is either suppressed or at its local maximum intensity with respect to the polarization angle. Accordingly, the rota-

tional angle ϕ to be considered in Table II is 0° (blue) or 90° (green) for parallel configuration, and 0° (orange) for crossed configuration. Thus, the symmetry of the observed phonon peaks can be directly determined on the basis of the selection rules given in Table II. Specifically, A_g modes appear on all faces in parallel but never in crossed configuration, with their intensities depending on the tensor elements a , b , and c . In contrast, B_{1g} and B_{3g} modes are exclusively visible in crossed configuration. B_{1g} modes appear on the (001) and (101) faces, whereas B_{3g} modes are found on the (100) and (101) faces. On the (101) face, the corresponding intensity is reduced by approximately a factor of 2 compared to the (001) and (100) faces, respectively. B_{2g} modes are visible only on the (010) face in crossed configuration and on the (101) face in parallel- 90° configuration, with nearly equal intensities in both cases. Only peaks that satisfy these conditions are considered as first-order Raman-active modes. Additional weak peaks are disregarded, which arise from a slight misalignment of up to $\pm 3.3^\circ$ between the crystal and laboratory coordinate system in the XY plane, from the finite efficiencies of the polarizers and HWP’s, or from the nonzero NA of the used objective. By careful analysis of the spectra in Fig. 1, one can identify 19 of the 24 active Raman modes, of which many overlap and are indistinguishable in unpolarized measurements. The positions of all observed modes are indicated by solid, dashed, dash-dotted, and dotted vertical lines for A_g , B_{1g} , B_{2g} , and B_{3g} symmetry, respectively, and are summarized in Table III. The remaining five modes are expected to be either obscured by a mode of the same symmetry or very weak in intensity and are discussed later with the introduction of DFT results.

Full scans of polarization-angle resolved measurements on the (101) plane are shown in Fig. 2 and in the Supplemental Material (SM) [31] for the other faces. For reference, the top panel displays the spectrum at $\phi = 200^\circ$ where most of the modes are visible (analogous to Fig. 1), while intensities at a fixed wave number ($\omega = 243 \text{ cm}^{-1}$, B_{2g}^3 mode) are plotted on the right for both parallel and crossed configurations. A rotational misalignment between the laboratory coordinate system and the crystal axes can be inferred from the systematic offsets of all peak maxima relative to their expected positions calculated from Table II, and was later quantified from the fit as -6.7° . Similarly, this offset is -20.9° (100), 11.7° (010), and -6.9° (001) for the other faces in the SM. In addition, the background is modeled as $\cos^2(2\phi)$ and $\sin^2(2\phi)$ for parallel and crossed polarization configuration, respectively, as it shows a faint angle dependent intensity. We consider this to be an artifact of the experimental setup, but the exact mechanism remains unclear. The red curves in Fig. 2 represent slices from the modeling results that were determined by the procedure described in the following paragraph.

To obtain the Raman tensor elements from polarization dependent Raman data, the common approach is to analyze each spectrum separately for a fixed geometry and polarization configuration at different rotation angles. Each spectrum is fitted with a set of line shapes, here Lorentz functions, each corresponding to a specific phonon mode. From the fitting results, the amplitudes, i.e., integrated peak areas, are extracted and fitted with the intensity profiles given by Eq. (2). However, this method becomes unreliable when phonon modes are close in frequency, such as the A_g^2 and B_{1g}^1 ($\approx 130 \text{ cm}^{-1}$)

TABLE III. Frequencies of the Raman-active Γ -point phonons ω_p in cm^{-1} as well as relative Raman tensor elements divided by the square root of the Raman shift-dependent intensity prefactor $F(\omega_p)$ from Eq. (4) and normalized to 1000. The first row lists the experimental values obtained from the multidimensional fit. The second row (values in parentheses) contains the corresponding theoretical results (frequencies obtained from QUANTUM ESPRESSO and Raman tensor elements obtained from CRYSTAL23). For comparison with the experimental data, the theoretical Raman tensor elements were uniformly scaled to minimize the mean squared deviation from the experimental values.

Mode index	A_g				B_{1g}		B_{2g}		B_{3g}	
	ω_p	a	b	c	ω_p	d	ω_p	e	ω_p	f
1	104 (103)	66 (24)	52 (76)	-58 (-82)	135 (120)	76 (156)	107 (105)	-45 (-26)	121 (114)	132 (137)
2	131 (126)	-75 (-95)	155 (176)	-1 (0)	191 (180)	-461 (-540)	154 (157)	-265 (-256)	262 (244)	87 (98)
3	211 (206)	70 (-70)	609 (523)	-435 (-366)	319 (317)	487 (658)	243 (243)	556 (542)	397 (385)	606 (916)
4	274 (273)	701 (636)	-244 (-267)	-371 (-301)	- (402)	- (-109)	310 (303)	354 (359)	- (397)	- (-55)
5	362 (374)	-352 (-426)	-93 (-28)	993 (901)	- (553)	- (-4)	401 (394)	-576 (-535)	- (602)	- (77)
6	393 (386)	-699 (-638)	-509 (-418)	1000 (781)			484 (492)	105 (224)		
7	423 (423)	94 (-40)	-257 (-168)	788 (753)			- (597)	- (16)		

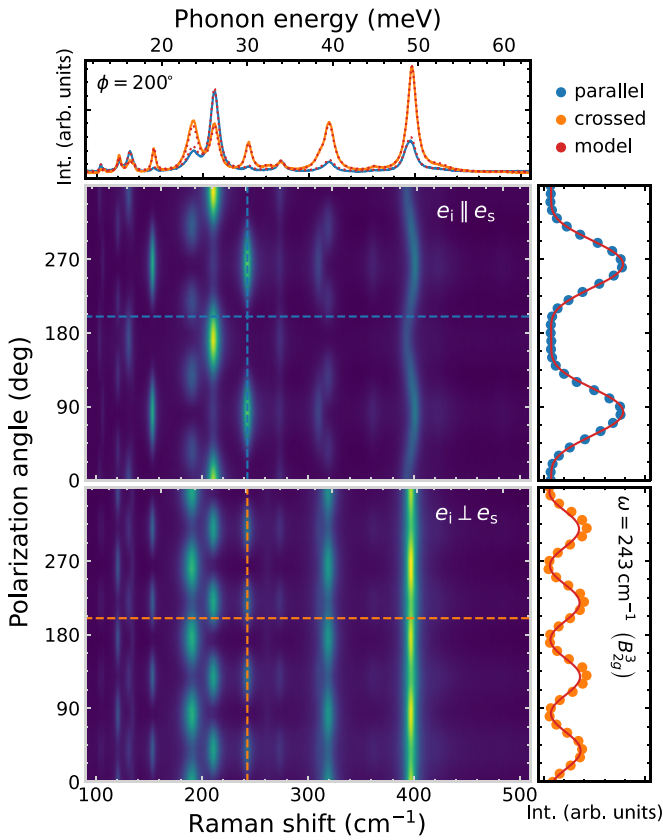


FIG. 2. Polarization-angle resolved Raman spectra of the (101) plane. The upper (lower) contour plot as well as blue (orange) lines and symbols correspond to measurements in parallel (crossed) polarization configuration. Red curves represent results from the hyperspectral fitting procedure. The panels above and to the right display slices of the data at 200° and 243 cm^{-1} (B_{2g}^3 mode), respectively.

or A_g^6 , B_{3g}^4 , and B_{2g}^5 ($\approx 400 \text{ cm}^{-1}$) modes, due to the strong correlation of the peak-describing parameters. To overcome these limitations, a multidimensional (hyperspectral) fitting procedure was employed, that incorporates every spectrum measured across every geometry, in both parallel and crossed polarization, and at all rotation angles in one single global fit. In this approach, the angle-dependent intensity profiles from Table II serve as modulation functions for each phonon line shape. This procedure constrains the evolution of the amplitude parameter and helps to separate overlapping modes by also exploiting information from multiple geometries and polarization configurations. More information about this method can be found in Ref. [32] and a descriptive demonstration in the SM [31]. The signs of the Raman tensor elements b , d , e , and f cannot be determined from the experiment as they appear only in squared form in the selection rules. In addition, birefringence allows the signs of a and c to be fixed only relative to one another, meaning they are either identical or opposite. By comparison with the theoretical results in Table III, we chose the sign to maximize the agreement with the calculated tensor elements but note that this assignment may be ambiguous for tensor elements whose values are close to zero. The fitted phonon frequencies and Raman tensor elements, divided by the square root of the Raman shift-dependent prefactor $F(\omega_p)$, are listed in Table III.

Figure 3 shows the DFT-calculated phonon dispersion along the high-symmetry path $\Gamma\text{ZUX}\Gamma\text{YTRSY|ZT|UR|XS}$. Additionally, the phonon density of states is depicted in the right panel of the figure. Owing to their masses, low-frequency modes are dominated by motions of the La atoms, followed by a broad distribution of In atoms. Modes with frequencies higher than $\approx 300 \text{ cm}^{-1}$ consist almost entirely of O vibrations. The obtained Γ point phonon energies, which

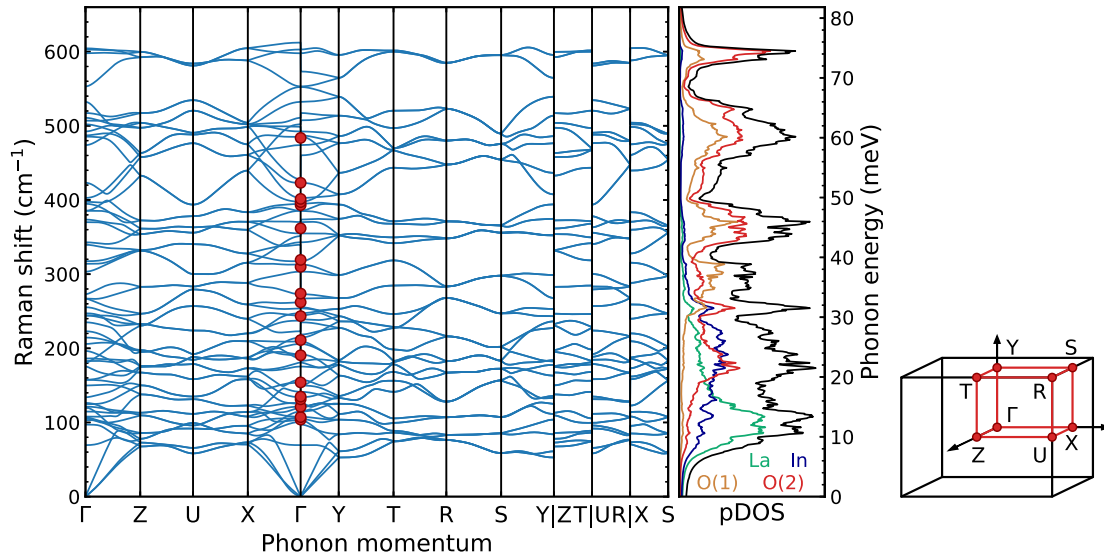


FIG. 3. Phonon dispersion of LIO along the high symmetry path displayed in the first Brillouin zone on the right side. Lines indicate DFT calculations and symbols mark experimentally obtained results. The right panel displays the site-resolved partial pDOS together with the total pDOS plotted in black.

are uniformly scaled by a factor of 1.023 as justified in Sec. III C, align very well with the experiment (Table III). The calculated Raman tensor elements, which are also shown in Table III, however, show stronger deviations between experimental and theoretical results with a mean absolute error of 7% relative to the maximum intensity.

Both DFT calculations further indicate that the modes most likely to remain unobserved experimentally are the B_{1g}^4 , B_{1g}^5 , B_{2g}^7 , B_{3g}^4 , and B_{3g}^5 modes. According to the atomic displacement patterns (see SM [31]), these modes are O dominated and possess strong stretchinglike character. Similar stretching modes have been reported to display weak or vanishing Raman intensities in perovskites that do not undergo Jahn-Teller distortions, i.e. symmetry breaking due to spatially degenerate electronic ground states, or at least have small deviations in their B-O bond lengths. Examples include CaMnO_3 and metallic $Pnma$ compounds [33]. Since the In^{3+} ions in LIO possess an electronically nondegenerate $4d^{10}$ electron configuration, they do not support a Jahn-Teller distortion, which provides a possible explanation for the absence of these modes in our measurements.

Consistently, the absolute values of the calculated Raman tensor elements for the missing modes (see Table III) are found to be small (≤ 109 in normalized units). We note that the intensity resulting from these low tensor elements would be further attenuated by the frequency-dependent prefactor in Eq. (4). In particular, a mode at 400 or 550 cm^{-1} would be reduced in intensity by approximately a factor of 3 or 4, respectively, relative to a mode at 100 cm^{-1} with identical Raman tensor elements.

Figure 4 shows the oscillation patterns of the B_{2g}^1 and B_{1g}^3 modes at the Γ point, with the remaining modes provided in the SM [31]. The B_{2g}^1 mode is dominated by oscillations of the La atoms, whereas the B_{1g}^3 mode exclusively involves O-site oscillations. Consequently, these two modes might serve as sensitive indicators to be analyzed for specific crystal modifications, such as alloying in $\text{La}_{(1-x)}\text{Ga}_x\text{InO}_3$ or doping

on O sites. Furthermore, the $Pnma$ space group of ABO_3 perovskites dictates specific characteristics of the atomic motions: Following the site symmetry of $4c$ ($C_s = \{E, m_{010}\}$), displacements of A and O(1) atoms are constrained to the XZ plane for A_g and B_{2g} modes, while B_{1g} and B_{3g} modes are confined along the Y direction. In contrast, there are no restrictions for the oscillations of O(2) atoms, as their site symmetry of $8d$ ($C_1 = \{E\}$) only contains the unit operation. In many perovskites, such as LaMnO_3 or YMnO_3 , however, the O(2) vibrations are nearly orientated either parallel or perpendicular to the B-O(2) bonds, which are, due to the shorter bonding lengths, significantly stronger than the A-O(2) bonds [34]. In LIO, the difference between these two bond types is smaller, resulting in less correlated O(2)-displacements. Although some modes correspond to nearly pure vibrations of an isolated BO_6 octahedron, including breathing (B_{3g}^5), tilting

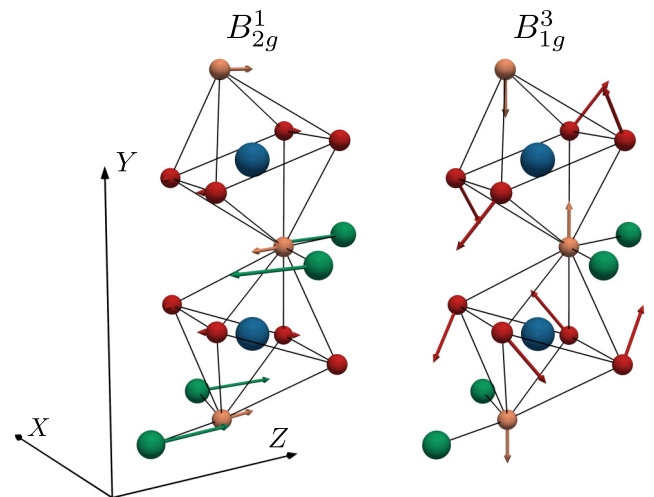


FIG. 4. Phonon displacement patterns of the B_{2g}^1 and B_{1g}^3 modes in LIO. La, In, O(1), and O(2) atoms are depicted in green, blue, orange, and red, respectively.

(A_g^4), bending (B_{3g}^2), and rotational (B_{1g}^2) modes, many of the displacement patterns represent mixtures of these motions and thus cannot be uniquely labeled, unlike in similar $Pnma$ perovskites.

V. SUMMARY

Summarizing and concluding, we studied the lattice dynamics of orthorhombic LIO using polarization-angle resolved Raman spectroscopy in combination with DFT calculations. Measurements in backscattering geometry on multiple crystallographic surfaces allowed the identification and symmetry assignment of 19 of the 24 Raman-active Γ -point phonon modes. We discuss the remaining modes and relate their absence in our spectra to their strong stretchinglike character. A multidimensional fitting approach was employed to analyze the angular dependence of the Raman scattering intensities and to extract the relative Raman tensor elements, even for closely spaced and overlapping modes. First-principles DFT calculations were used to compute the phonon dispersion, phonon density of states, and atomic displacement patterns. The calculated Γ -point phonon frequencies were found to be in good overall agreement with the experiment.

These results provide a reference for future studies addressing strain, alloying, or defect-induced modifications of the lattice, as well as for investigations of phonon-related transport and optical processes in LIO-based oxide heterostructures and electronic devices.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

- [1] N. Reyren, S. Thiel, A. D. Caviglia, L. F. Kourkoutis, G. Hammerl, C. Richter, C. W. Schneider, T. Kopp, A.-S. Rüetschi, D. Jaccard, M. Gabay, D. A. Muller, J.-M. Triscone, and J. Mannhart, Superconducting interfaces between insulating oxides, *Science* **317**, 1196 (2007).
- [2] A. Brinkman, M. Huijben, M. Van Zalk, J. Huijben, U. Zeitler, J. C. Maan, W. G. Van Der Wiel, G. Rijnders, D. H. A. Blank, and H. Hilgenkamp, Magnetic effects at the interface between non-magnetic oxides, *Nat. Mater.* **6**, 493 (2007).
- [3] A. Ohtomo and H. Y. Hwang, A high-mobility electron gas at the $\text{LaAlO}_3/\text{SrTiO}_3$ heterointerface, *Nature (London)* **427**, 423 (2004).
- [4] H. J. Kim, U. Kim, H. M. Kim, T. H. Kim, H. S. Mun, B.-G. Jeon, K. T. Hong, W.-J. Lee, C. Ju, K. H. Kim, and K. Char, High mobility in a stable transparent perovskite oxide, *Appl. Phys. Express* **5**, 061102 (2012).
- [5] H. J. Kim, U. Kim, T. H. Kim, J. Kim, H. M. Kim, B.-G. Jeon, W.-J. Lee, H. S. Mun, K. T. Hong, J. Yu, K. Char, and K. H. Kim, Physical properties of transparent perovskite oxides (Ba, La) SnO_3 with high electrical mobility at room temperature, *Phys. Rev. B* **86**, 165205 (2012).
- [6] K. S. Belthle, U. N. Gries, M. P. Mueller, D. Kemp, A. Prakash, M. Rose, J. M. Börgers, B. Jalan, F. Gunkel, and R. A. De Souza, Quantitative determination of native point-defect concentrations at the ppm level in un-doped BaSnO_3 thin films, *Adv. Funct. Mater.* **32**, 2113023 (2022).
- [7] U. Kim, C. Park, T. Ha, Y. M. Kim, N. Kim, C. Ju, J. Park, J. Yu, J. H. Kim, and K. Char, All-perovskite transparent high mobility field effect using epitaxial BaSnO_3 and LaInO_3 , *APL Mater.* **3**, 036101 (2015).
- [8] Y. Kim, Y. M. Kim, J. Shin, and K. Char, $\text{LaInO}_3/\text{BaSnO}_3$ polar interface on MgO substrates, *APL Mater.* **6**, 096104 (2018).
- [9] H. Paik, Z. Chen, E. Lochocki, H. A. Seidner, A. Verma, N. Tanen, J. Park, M. Uchida, S. Shang, B.-C. Zhou, M. Brützmann, R. Uecker, Z.-K. Liu, D. Jena, K. M. Shen, D. A. Muller, and D. G. Schlom, Adsorption-controlled growth of La-doped BaSnO_3 by molecular-beam epitaxy, *APL Mater.* **5**, 116107 (2017).
- [10] G. Hoffmann, M. Zupancic, A. A. Riaz, C. Kalha, C. Schlueter, A. Gloskovskii, A. Regoutz, M. Albrecht, J. Nordlander, and O. Bierwagen, Enabling 2D electron gas with high room-temperature electron mobility exceeding $100 \text{ cm}^2 \text{ V s}^{-1}$ at a perovskite oxide interface, *Adv. Mater.* **36**, 2409076 (2024).
- [11] D. Pfützenreuter, M. Zupancic, Z. Galazka, R. Schewski, A. Dittmar, K. Irmscher, M. Albrecht, and J. Schwarzkopf, Epitaxial BaSnO_3 thin films with low dislocation density grown on lattice matched LaInO_3 substrates, *Nanotechnology* **32**, 505609 (2021).
- [12] Z. Galazka, K. Irmscher, S. Ganschow, M. Zupancic, W. Aggoune, C. Draxl, M. Albrecht, D. Klimm, A. Kwasniewski, T. Schulz, M. Pietsch, A. Dittmar, R. Grueneberg, U. Juda, R. Schewski, S. Bergmann, H. Cho, K. Char, T. Schroeder, and M. Bickermann, Melt growth and physical properties of bulk LaInO_3 single crystals, *Phys. Status Solidi A* **218**, 2100016 (2021).
- [13] W. Aggoune and C. Draxl, Tuning two-dimensional electron and hole gases at $\text{LaInO}_3/\text{BaSnO}_3$ interfaces by polar distortions, termination, and thickness, *npj Comput. Mater.* **7**, 174 (2021).
- [14] H. He, X. Huang, and L. Chen, The effects of dopant valence on the structure and electrical conductivity of LaInO_3 , *Electrochim. Acta* **46**, 2871 (2001).
- [15] S. Kumar, G. Dwivedi, A. G. Joshi, S. Chatterjee, and A. Ghosh, Study of structural, dielectric, optical properties and electronic

- structure of Cr-doped LaInO_3 perovskite nanoparticles, *Mater. Charact.* **131**, 108 (2017).
- [16] M. I. Aroyo, J. M. Perez-Mato, C. Capillas, E. Kroumova, S. Ivantchev, G. Madariaga, A. Kirov, and H. Wondratschek, Bilbao crystallographic server: I. Databases and crystallographic computing programs, *Z. Kristallogr.-Cryst. Mater.* **221**, 15 (2006).
- [17] E. Kroumova, M. Aroyo, J. Perez-Mato, A. Kirov, C. Capillas, S. Ivantchev, and H. Wondratschek, Bilbao crystallographic server: Useful databases and tools for phase-transition studies, *Phase Trans.* **76**, 155 (2003).
- [18] C. Kranert, C. Sturm, R. Schmidt-Grund, and M. Grundmann, Raman tensor formalism for optically anisotropic crystals, *Phys. Rev. Lett.* **116**, 127401 (2016).
- [19] C. Kranert, C. Sturm, R. Schmidt-Grund, and M. Grundmann, Raman tensor elements of $\beta\text{-Ga}_2\text{O}_3$, *Sci. Rep.* **6**, 35964 (2016).
- [20] W. Hayes and R. Loudon, *Scattering of Light by Crystals* (Wiley, New York, 1978), p. 31.
- [21] H. B. Ribeiro, M. A. Pimenta, C. J. S. De Matos, R. L. Moreira, A. S. Rodin, J. D. Zapata, E. A. T. De Souza, and A. H. Castro Neto, Unusual angular dependence of the Raman response in black phosphorus, *ACS Nano* **9**, 4270 (2015).
- [22] T. Strach, J. Brunen, B. Lederle, J. Zegenhagen, and M. Cardona, Determination of the phase difference between the Raman tensor elements of the A_{1g} -like phonons in $\text{SmBa}_2\text{Cu}_3\text{O}_{7-\delta}$, *Phys. Rev. B* **57**, 1292 (1998).
- [23] M. Zupancic, W. Aggoune, T. Markurt, Y. Kim, Y. M. Kim, K. Char, C. Draxl, and M. Albrecht, Role of the interface in controlling the epitaxial relationship between orthorhombic LaInO_3 and cubic BaSnO_3 , *Phys. Rev. Mater.* **4**, 123605 (2020).
- [24] P. Giannozzi, S. Baroni, N. Bonini, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, G. L. Chiarotti, M. Cococcioni, I. Dabo, A. Dal Corso, S. De Gironcoli, S. Fabris, G. Fratesi, R. Gebauer, U. Gerstmann, C. Gougoussis, A. Kokalj, M. Lazzeri, L. Martin-Samos, *et al.*, QUANTUM ESPRESSO: A modular and open-source software project for quantum simulations of materials, *J. Phys.: Condens. Matter* **21**, 395502 (2009).
- [25] G. Prandini, A. Marrazzo, I. E. Castelli, N. Mounet, and N. Marzari, Precision and efficiency in solid-state pseudopotential calculations, *npj Comput. Mater.* **4**, 72 (2018).
- [26] P. Hartley, R. G. Egdel, K. H. L. Zhang, M. V. Hohmann, L. F. J. Piper, D. J. Morgan, D. O. Scanlon, B. A. D. Williamson, and A. Regoutz, Experimental and theoretical study of the electronic structures of lanthanide indium perovskites LnInO_3 , *J. Phys. Chem. C* **125**, 6387 (2021).
- [27] J. M. Skelton, D. Tiana, S. C. Parker, A. Togo, I. Tanaka, and A. Walsh, Influence of the exchange-correlation functional on the quasi-harmonic lattice dynamics of II-VI semiconductors, *J. Chem. Phys.* **143**, 064710 (2015).
- [28] A. Erba, J. K. Desmarais, S. Casassa, B. Civalleri, L. Donà, I. J. Bush, B. Searle, L. Maschio, L. Edith-Daga, A. Cossard, C. Ribaldone, E. Ascrizzi, N. L. Marana, J.-P. Flament, and B. Kirtman, CRYSTAL23: A program for computational solid state physics and chemistry, *J. Chem. Theory Comput.* **19**, 6891 (2023).
- [29] L. Schimka, J. Harl, and G. Kresse, Improved hybrid functional for solids: The HSEsol functional, *J. Chem. Phys.* **134**, 024116 (2011).
- [30] J. Laun and T. Bredow, BSSE-corrected consistent Gaussian basis sets of triple-zeta valence with polarization quality of the fifth period for solid-state calculations, *J. Comput. Chem.* **43**, 839 (2022).
- [31] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/frdm-2plg> for a detailed description of an exemplary calculation of the angle dependent selection rules, all Raman spectra on the planes not shown in the main text, details on the reliability of the fit, and all 24 displacement patterns of the Raman-active phonon modes.
- [32] H. Tornatzky, J. Rose, M. Meißner, B. M. Janzen, Z. Galazka, J. S. Reparaz, M. E. Ramsteiner, and M. R. Wagner, Complete Raman tensor determination in birefringent $\beta\text{-Ga}_2\text{O}_3$ by single-stage hyperspectral analysis of polarization angle-resolved Raman spectra, [arXiv:2607.21045](https://arxiv.org/abs/2607.21045).
- [33] L. Martín-Carrón, A. de Andrés, M. J. Martínez-Lope, M. T. Casais, and J. A. Alonso, Raman phonons as a probe of disorder, fluctuations, and local structure in doped and undoped orthorhombic and rhombohedral manganites, *Phys. Rev. B* **66**, 174303 (2002).
- [34] M. N. Iliev, M. V. Abrashev, H.-G. Lee, V. N. Popov, Y. Y. Sun, C. Thomsen, R. L. Meng, and C. W. Chu, Raman spectroscopy of orthorhombic perovskitelike YMnO_3 and LaMnO_3 , *Phys. Rev. B* **57**, 2872 (1998).