Mechanism of unusual elasticity in β-Ga_2O_3 studied by ab-initio calculation

第一原理計算を用いた酸化ガリウムの異常弾性機構の解明

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1. Introduction

The study of β-Ga_2O_3 dates back over 100 years (in 1925), [1] and a lot of studies on its material properties were reported until now, including detailed crystallographic structure, thermal conductivity, and electron mobility. However, the elastic constants C_{ij} of β-Ga_2O_3 have not been reported at all for nearly a century. This fact is very surprising for the following reasons: (1) C_{ij} are one of the most fundamental properties to be aggressively determined in scientific studies because they provide information about the curvature of interatomic potential. (2) β-Ga_2O_3 is a promising candidate for next-generation power devices because of the second highest bandgap among semiconductors and low manufacturing cost. The elastic properties are essential in designing a β-Ga_2O_3-based power device because they are needed to evaluate stresses caused by difference in lattice parameter and thermal expansion from another contacting material. Thus, the clarification of β-Ga_2O_3's elasticity is significant challenge both for scientific and practical studies.

The monoclinic β-Ga_2O_3 crystal belongs to space group *C2/m* and has a polyhedral-linkage structure in which tetrahedra and octahedra composed of gallium and oxygen atoms are joined by oxygen atoms as shown in Fig. 1. The elastic-constant matrix is expressed as follows:

$$[C_{ij}] = \begin{bmatrix} C_{11} & C_{12} & C_{13} & 0 & C_{15} & 0 \\ & C_{22} & C_{23} & 0 & C_{25} & 0 \\ & & C_{33} & 0 & C_{35} & 0 \\ & & & C_{44} & 0 & C_{46} \\ & sym. & & & C_{55} & 0 \\ & & & & & C_{66} \end{bmatrix} \quad (1)$$

We have determined all the thirteen components of β-Ga_2O_3 with high precision using resonant ultrasound spectroscopy coupled with laser-Doppler interferometry. [2] The method unambiguously identifies resonance frequencies by comparing measured and calculated displacement distributions, leading to the accurate determination of C_{ij} with inverse calculation. [3] The measurements of C_{ij} revealed the unusual elastic properties of β-Ga_2O_3, including strong anisotropies in the longitudinal (C_{11} << C_{22}, C_{33}) and shear (C_{44} << C_{55}, C_{66}) moduli, and Poisson's ratio exceeding 0.5.

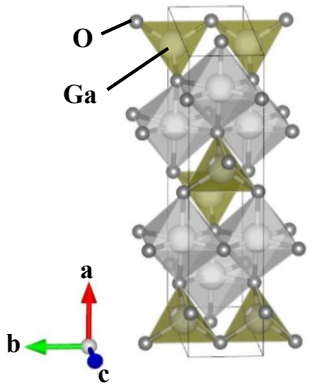

Fig. 1 The conventional unit cell of β-Ga_2O_3.

The present study clarifies the mechanism of these elastic properties with ab-initio calculation; the computed C_{ij} are essentially consistent with our measurements and reproduce the anomalous properties. The deformation behavior of a β-Ga_2O_3 crystal is also reproduced by first-principle calculation, revealing that the unusual elasticity in β-Ga_2O_3 results from the truss-like deformation of its specific network structure composed of tetrahedra and octahedra.

2. Calculation procedure

In the theoretical study, we utilized the Vienna ab-initio simulation package (VASP). The exchange correlation potentials are expressed by the local density approximation (LDA) and the generalized gradient approximation (GGA). We used the plane-waves cutoff energy of 1,300 eV and 10×10×10 mesh k points. We applied thirteen different deformation modes to the unit cell up to ± 1 % in each deformation mode, and calculated the total energy vs. strain curve. C_{ij} are obtained by fitting a harmonic function to the curve. [4] We also calculated the atomic migration in β-Ga_2O_3 caused by the simple tensile or shear strain of 1 % using the LDA potential. The atoms inside the cell are relaxed in all calculation.

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3. Results and Discussion

The calculation with LDA provides both the lattice parameters and the diagonal elastic constants closer to measurements than with GGA, indicating that the LDA potential is preferable in predicting material properties of β-Ga_2O_3 with first-principle calculation.

The unusual elastic properties of β-Ga_2O_3 are also shown in theoretical calculation. We attribute them to the truss-like polyhedral-linkage crystallographic structure. Its projection views onto principal planes are demonstrated in Fig. 2, where the rigid tetrahedra and octahedra are connected by sharing the vertex or side. β-Ga_2O_3 can produce an apparent large longitudinal deformation along the x_1 axis with less Ga-O bond distance change in the rigid polyhedral by the bending of connection between the tetrahedron and octahedron. Furthermore, the change of the square basal plane of octahedra into a parallelogram allows shear deformation in the x_2-x_3 plane with less angle change of the rigid O-Ga-O bond. These crystallographic properties result in remarkably small C_{11} and C_{44}, that is, significant elastic anisotropies. To check the validity of our interpretation, we calculated the atomic behavior in the octahedron when the simple tensile or shear strain, corresponding to the diagonal elastic constants, is applied. Table I shows the change ratios of the neighboring Ga-O bond distance and the O-Ga-O bond angle. As can be seen from Table I, the bond distance change required to produce the 1 % tensile strain along the x_1 axis is less than along the x_2 and x_3 axis by ~18 %; and it is possible to produce equivalent shear strain in the x_2-x_3 plane through the smaller bond angle change than the x_1-x_2 plane by ~70 %, supporting our view.

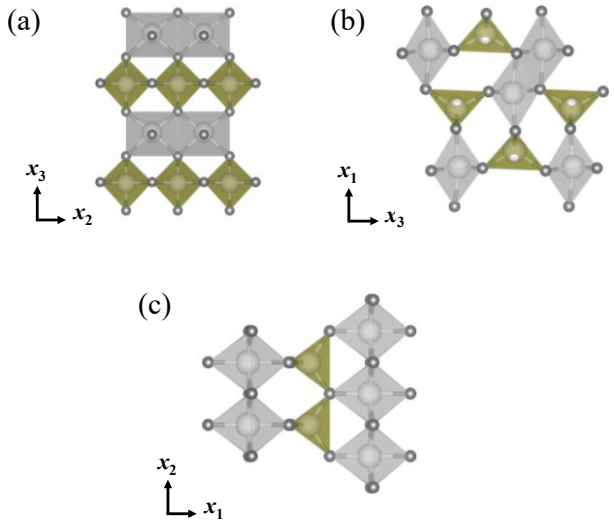

Fig. 2 Schematic projection views of the crystal structure on the (a) x_2-x_3, (b) x_1-x_3, and (c) x_1-x_2 planes.

Table I. Changes in the neighboring Ga-O bond distance and the O-Ga-O bond angle in the octahedron caused by the simple tensile or shear strain of 1 %.

	Change ratio (%)	
strain	distance	angle
ε_{11}	0.276	0.289
ε_{22}	0.382	0.412
ε_{33}	0.292	0.255
$2\varepsilon_{23}$	0.374	0.196
$2\varepsilon_{31}$	0.056	0.562
$2\varepsilon_{12}$	0.193	0.656

Finally, we consider anomalous Poisson's ratio: ν_{21} and ν_{31} exceed 0.5. β-Ga_2O_3 has a truss-like structure in the x_1-x_3 and x_1-x_2 planes, where two tetrahedra and two octahedra form parallelogram-shape units. They function as square truss, providing larger Poisson's ratio in these plane. Besides, the transverse strain caused by the uniaxial stress applied along the x_2 or x_3 axis becomes larger in the x_1 direction because of the smaller C_{11}, so that ν_{21} and ν_{31} become larger. For these reasons, ν_{21} and ν_{31} can exceed 0.5.

4. Conclusion

We theoretically studied the unusual elasticity of β-Ga_2O_3 by calculating elastic constants and atomic behavior caused by principal strains through ab-initio calculation. The calculated values with LDA for lattice parameters and elastic constants show good agreements with the measurements, indicating that it would be better to use the LDA potential in predicting other properties with this calculation method. We explained the unusual elastic properties of β-Ga_2O_3 with the truss-like deformation of the polyhedral-linkage structure and confirmed this consideration by first-principle calculation.

References

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