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研究課題名

THz 波吸収特性の制御のための FAPbBr<sub>x</sub>I<sub>3-x</sub> ハイブリッドペロブスカイトの欠陥

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<研究内容・成果等の要約>

From this research grant, we have obtained several important results and published 4 papers as below.

1. Finding of the correlation of THz-wave absorption property by different halogen elements in FAPb(Br, I)-based hybrid perovskite thin films
2. Understanding of the bias effect on surface chemical states of CH<sub>3</sub>NH<sub>3</sub>PbBr<sub>3</sub> hybrid perovskite single crystal and finding of the method for decreasing CH<sub>3</sub>NH<sub>2</sub> molecular defect
3. Understanding of THz wave absorption property of all mixed organic-inorganic hybrid perovskite thin film, MA(Sn, Pb)(Br, I)<sub>3</sub>
4. Finding and understanding of unusual THz-wave absorptions in  $\delta/r$ -mixed- phase FAPbI<sub>3</sub> single crystals: interfacial phonon vibration modes

From this research, we confirmed that

1. Element dependence of THz-wave absorption by the metal cations and halogen anions such as Sn/Pb and Br/I, respectively
2. Creation of the new phonon mode in mixed phases induced with the interfacial vibration

Now, we are trying to make a real THz-based device such as sensor, detector, and modulator. We believe these results with the suggested fabrication method and physical properties will support deeply to realize a THz device.

<研究発表（口頭、ポスター、誌上別）>

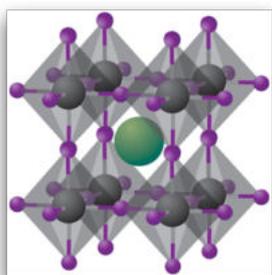
1. Oral presentation (Invited)

- (1) “Defect and its property on organic-inorganic hybrid perovskite: First step for THz-application research”, **Min-Cherl Jung**, Invited seminar, Materials Genome Institute, Shanghai University, China, Nov. 2020

2. Publications (Peer-review)

- (1) Inhee Maeng, Seungjun Lee, E. Q. Han, Yurou Zhang, Seung Jae Oh, Masakazu Nakamura, Jung-Ho Yun, Lianzhou Wang, Young-Kyun Kwon, and **Min-Cherl Jung (Corr. Author)**, "Unusual terahertz-wave absorptions in  $\delta/\alpha$ -mixed-phase FAPbI<sub>3</sub> single crystals: interfacial phonon vibration modes", **NPG Asia Materials**, **13**, 75 (2021)
- (2) Inhee Maeng, Asuka Matsuyama, Masakazu Nakamura, and **Min-Cherl Jung (Corr. Author)**, "Correlation of THz-wave absorption properties by different halogen elements in FAPb(Br, I)-based hybrid perovskite thin films", **Applied Physics Express**, **14**, 121002 (2021)
- (3) Inhee Maeng, Hiroshi Tanaka, Valynn Katrine Mag-usara, Makoto Nakajima, Masakazu Nakamura and **Min-Cherl Jung (Corr. Author)**, “Terahertz Wave Absorption Property of all Mixed Organic-Inorganic Hybrid Perovskite Thin Film MA(Sn, Pb)(Br, I)<sub>3</sub> Fabricated by Sequential Vacuum Evaporation Method”, **Frontiers in Chemistry**, **9**, 753141 (2021)
- (4) Young Mi Lee, Inhee Maeng, Miaoqiang Lyu, Jung-Ho, Yun, Lianzhou Wang, Masakazu Nakamura, and **Min-Cherl Jung (Corr. Author)**, “Bias effect on surface chemical states of CH<sub>3</sub>NH<sub>3</sub>PbBr<sub>3</sub> hybrid perovskite single crystal: Decreasing CH<sub>3</sub>NH<sub>2</sub> molecular effect”, **Applied Surface Science**, **542**, 148536 (2021)

<研究の目的、経過、結果、考察（5000 字程度、中間報告は2000 字程度）>



AMX<sub>3</sub> perovskite structure  
A: Green (Organic or inorganic cations)  
M: Gray (Metal cations)  
X: Purple (Halogen anion)

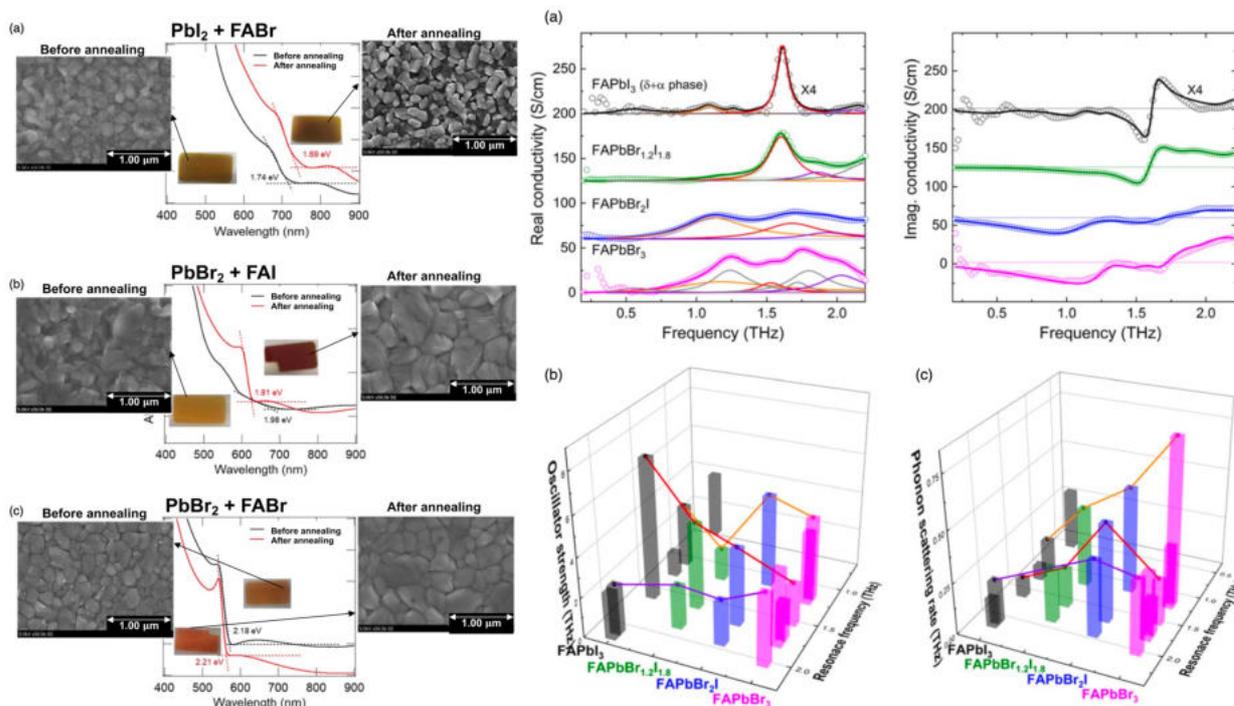
Figure 1. Typical organic-inorganic hybrid perovskite structure

Recently, organic-inorganic hybrid perovskite (OHP) materials, ABX<sub>3</sub> (A = Organic cation: CH<sub>3</sub>NH<sub>3</sub><sup>+</sup>(MA)/NH<sub>2</sub>CH=NH<sub>2</sub><sup>+</sup>(FA), B = Metal cation: Pb/Sn, and X = Halogen anion: Cl/Br/I) are the potential application material for solar cell, field-effect transistor, and light-emitting diode applications. OHP materials have shown amazing results, especially in case of solar cells, including a power conversion efficiency over 25 %, because of their key physical properties such as high absorption coefficient, high carrier mobility, and long carrier lifetime.

However, its fundamental physical properties such as defect, phonon-dispersion, and electronic structure are not fully understood yet. Interestingly, there is no serious research for the THz-wave absorption property (0.5-2.5 THz range) and its application possibility such as THz-wave modulation and sensing devices. In general assumption, the OHP has two cations and one anion in its perovskite structure that have various vibration modes such as metal cation-halogen anion and molecular vibrations in the range of 0.5 to 2.5 THz. (Fig. 1) To understand and control this physical property, the OHP material should be performed with the studies of defect, phonon-dispersion, and electronic structures. From these studies, we can expect that the device realization with a flexible and compacted THz-wave modulating/sensing properties will be come.

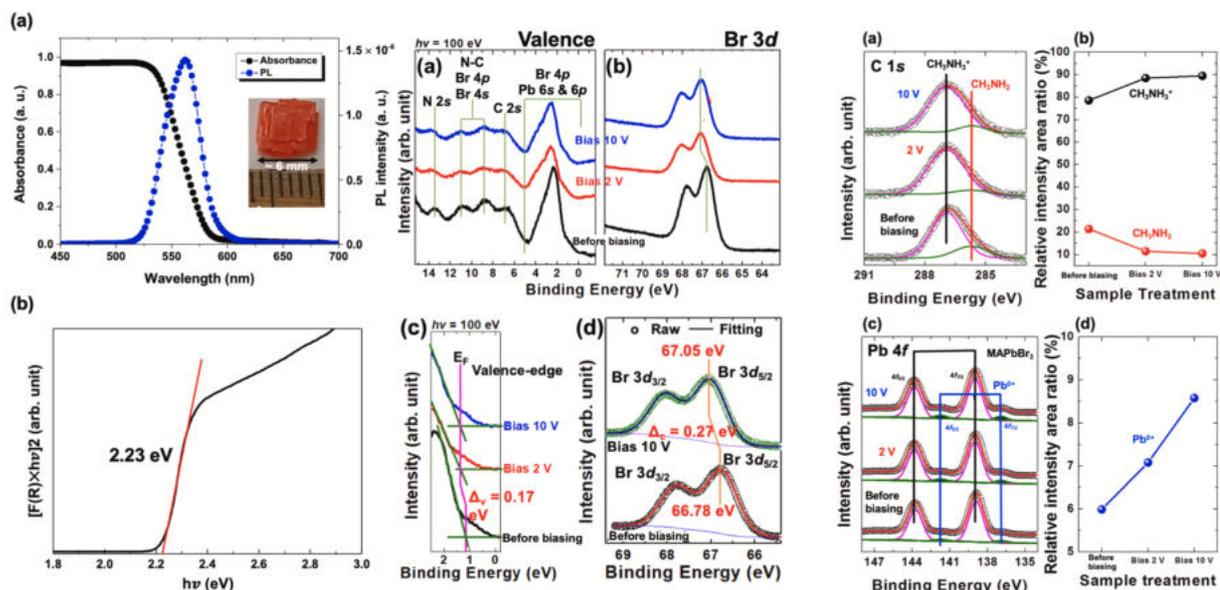
In this research supported by Izumi Zaidan (2020), we performed several experiments and obtained four important results to realize a THz-based device as below.

1. Finding of the correlation of THz-wave absorption property by different halogen elements in FAPb(Br, I)-based hybrid perovskite thin films



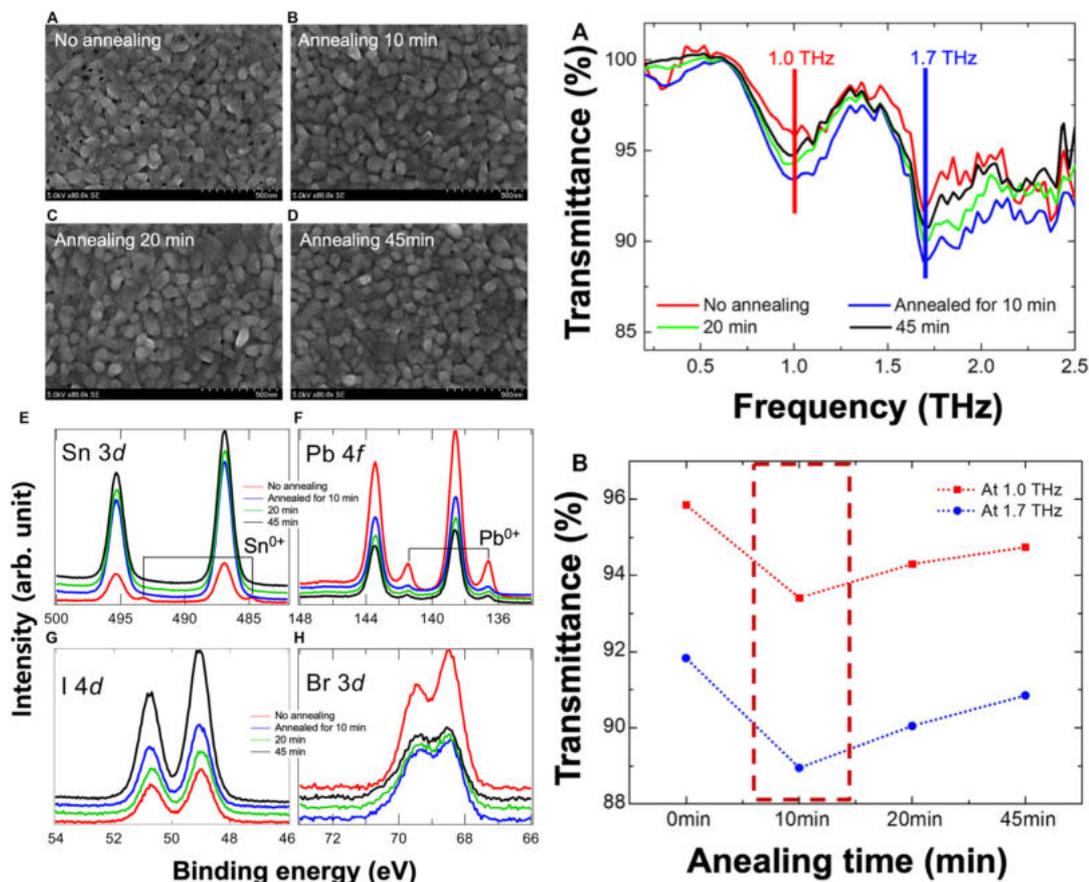
We confirmed the surface morphologies and THz-wave absorption properties in each thin film fabricated by the sequential vacuum evaporation.

2. Understanding of the bias effect on surface chemical states of  $\text{CH}_3\text{NH}_3\text{PbBr}_3$  hybrid perovskite single crystal and finding of the method for decreasing  $\text{CH}_3\text{NH}_2$  molecular defect



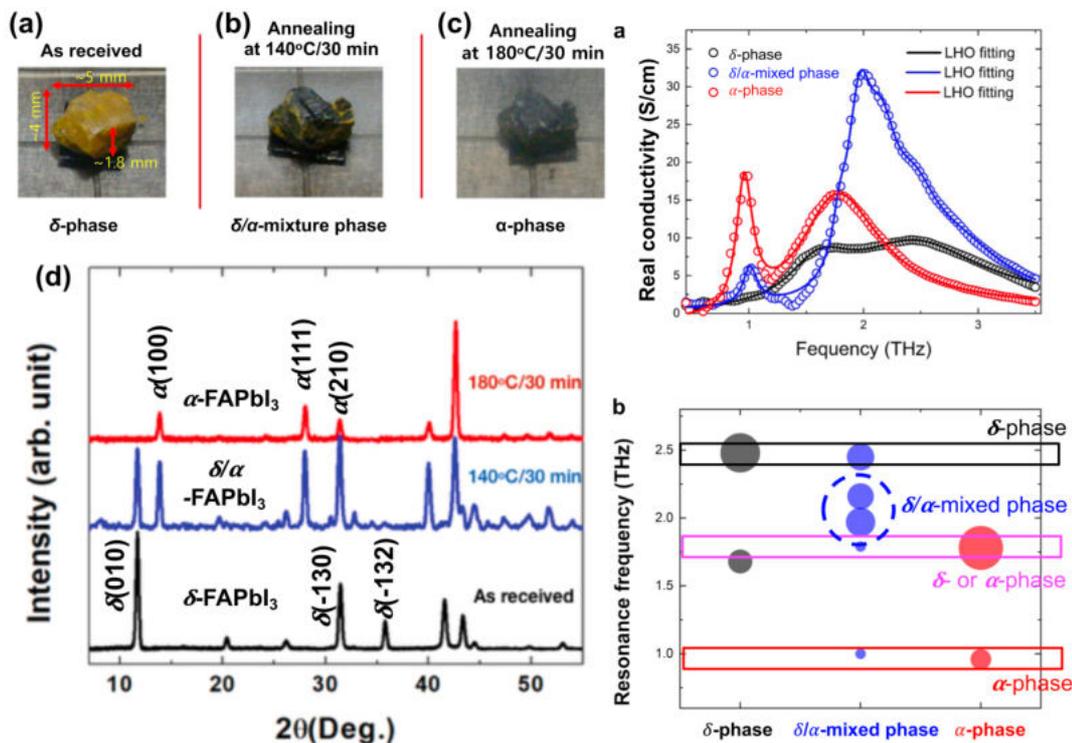
We confirmed the bias effect in organic-inorganic hybrid perovskite material and suggested the method to remove the molecular defect.

3. Understanding of THz wave absorption property of all mixed organic-inorganic hybrid perovskite thin film,  $\text{MA}(\text{Sn}, \text{Pb})(\text{Br}, \text{I})_3$

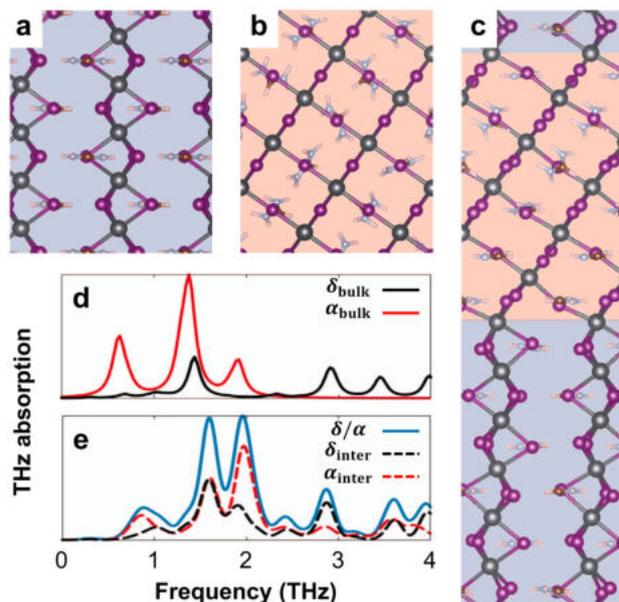


We fabricated all mixed organic-inorganic hybrid perovskite thin films by the sequential vacuum evaporation method and found the correlations among metal cations and halogen anions.

4. Finding and understanding of unusual THz-wave absorptions in  $\delta/\alpha$ -mixed-phase FAPbI<sub>3</sub> single crystals: interfacial phonon vibration modes



We found the new phonon mode in the THz range from 0.5 to 3 THz in FAPbI<sub>3</sub> single crystal and suggested the interfacial vibration mode in the mixed phase.



Also, we confirmed that

1. Element dependence of THz-wave absorption by the metal cations and halogen anions such as Sn/Pb and Br/I, respectively
2. Creation of the new phonon mode in mixed phases induced with the interfacial vibration

From these results, we are trying to make a real THz-based device such as sensor, detector, and modulator.