

## Ferroelectricity, Piezoelectricity, and Unprecedented Starry Ferroelastic Patterns in Organic–Inorganic $(\text{CH}_3\text{C}(\text{NH}_2)_2)_3[\text{Sb}_2\text{X}_9]$ ( $\text{X} = \text{Cl}/\text{Br}/\text{I}$ ) Hybrids

### Autorzy

Aleksandra Krupińska

Bogumiła Burzyńska

Vasyl Kinzhybalo

Błażej Dziuk

Przemysław Szklarz

Dariusz Kajewski

Jan K. Zaręba

Ada Drwęcka

Szymon J. Zelewski

Piotr Durlak

Piotr Zieliński

Paweł Sobieszczyk

Ryszard Jakubas

Anna Piecha-Bisiorek

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### Streszczenie

In this study, we present a novel class of lead-free hybrid antimony halides incorporating the acetamidinium cation, with the chemical compositions:  $(\text{CH}_3\text{C}(\text{NH}_2)_2)_3[\text{Sb}_2\text{Cl}_9]$  (**ACA**),  $(\text{CH}_3\text{C}(\text{NH}_2)_2)_3[\text{Sb}_2\text{Br}_9]$  (**ABA**), and  $(\text{CH}_3\text{C}(\text{NH}_2)_2)_3[\text{Sb}_2\text{I}_9]$  (**AIA**). Despite their seemingly analogous chemical formulations, these compounds exhibit diverse physical characteristics, predominantly dictated by the differences in their metal-halide architectures. Indeed, the anionic frameworks of **ACA** and **AIA** are reminiscent of well-known ferroelectric materials, with **ACA** distinguished by its piezoelectric and ferroelastic characteristics, underpinned by a buckled honeycomb two-dimensional (2D) layers of antimony chloride. Conversely, **AIA** is characterized by its ferroelectric attribute, with discrete bioctahedral units forming a zero-dimensional (0D) structure. A surprising structural deviation constitutes the anionic sublattice of **ABA**, which amalgamates features from both **ACA** and **AIA**, yielding an unprecedented hybrid two-component (0D + 2D) anionic architecture. The ferroelectric properties of **AIA** have been demonstrated through pyroelectric current measurements and hysteresis loop analyses. Additionally, the noncentrosymmetric nature of **ACA** and **AIA** has been corroborated by second harmonic generation experiments. The piezoelectricity of **ACA** was confirmed using piezoresponse force microscopy (PFM). Furthermore, observations under a polarizing microscope revealed distinct ferroelastic properties in both **ACA** and **ABA**, characterized by well-defined and abundant star patterns previously observed only in simple oxides and alloys.

### Słowa kluczowe

Cations, Crystal structure, Crystals, Nonlinear optics, Polarization

Kolekcja

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