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In their recent Article, Liu et al.¹ conclude that "Our results clarify that previous claims on ferroelectricity are, in fact, misguided by the elastic variation between the domains, rather than by true piezoelectric (electromechanical) effects, which suggests the twin domains are non-ferroelectric". However, their underlying interpretation of the band excitation frequency tracking results is flawed, and their presented data can well be explained by ferroelectric sample polarization.

A common method for mapping the local polarization in ferroelectric domains is piezoresponse force microscopy (PFM) where an a.c. voltage is applied between the bottom electrode of the sample and the cantilever tip of the atomic force microscope. The sample cantilever system can be considered as a driven damped harmonic oscillator with a resonance frequency f_0 . Liu et al. build their argument on the hypothesis that the amplitude and phase contrast of domains observed in single-frequency PFM (sf-PFM) originate from shifts in the contact resonance frequency due to different elastic moduli of neighbouring domains. To verify their hypothesis, they claim to observe a resonance frequency contrast between neighbouring domains in their Fig. 1b,c. Yet, a more detailed analysis of their measurement data questions this claim. First, the authors do not provide data on the crystal orientation of each grain's surface, which is mandatory to discuss surface effects originating from ferroelastic twinning and ionic charging. Second, in contrast to the earlier reports on domains in CH₃NH₃PbI₃ referenced in their paper, blurring and significant amounts of scanning artefacts do hamper the identification of domains. In particular, domains and domain walls on the grain that is highlighted in their Fig. 1b are hardly distinguishable, which renders a reliable interpretation of the presented data very difficult. Third, they have disregarded important own data towards the interpretation of their measurements. In the very same image (their Fig. 1b,c), on the grain in the lower right corner, only contrast between domains and domain walls is visible, and there is clearly no contrast between the domains themselves. This missing resonance frequency contrast implies equal elastic moduli of neighbouring domains and therefore conflicts with their central claim.

Liu et al. further allege that, except for the manuscript of Huang et al.², all PFM data in previous publications were acquired with sf-PFM and therefore would be susceptible to misinterpretation. We agree that the interpretation of data from resonance-enhanced PFM requires care. However, Vorpahl et al.³ performed dual-resonance tracking PFM and obtained similar results to previous sf-PFM data⁴. Thus, if resonance-enhanced PFM measurements are carried out carefully, they do well allow the identification of ferroelectric domains, in particular when horizontal and vertical piezoresponse components are distinguished.

Most importantly, ferroelectricity can very well be an explanation for the inhomogeneous ion distribution reported by the Liu et al. in their Fig. 4. In a ferroelectric crystal with mobile ions, the ions must contribute to the screening of the ferroelectric polarization. In a grain with out-of-plane polarization components, mobile ions would therefore accumulate at the sample surface. Polarization-induced surface charge differences may exert forces on the cantilever that could explain small changes in the resonance frequency of the cantilever sample system. For example, their Supplementary Fig. 4 could be interpreted as a contrast in resonance frequency of neighbouring domains, which may originate from out-of-plane polarization components in a ferroelectric crystal³. This is not discussed in the manuscript.

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Although the authors have used interesting and very advanced characterization techniques, which may be used in the future to gain further insight into the dynamics of ions in perovskite thin films, they have not presented conclusive evidence for the non-ferroelectricity of $CH_3NH_3PbI_3$ as stated in the conclusion of their paper. On the contrary, some of their measurement data are better described by ferroelectric crystal polarization, which would also be in accordance with earlier reports^{2–4}.

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Author contributions

All authors contributed to the analysis and interpretation of the literature that is discussed and jointly wrote the article.

Competing interests

The authors declare no competing interests.

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