Non-Ising and Chiral Ferroelectric Domain Walls: Insights from Nonlinear Optical Microscopy

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The properties of ferroelectric domain walls can significantly differ from those of their parent material. Elucidating their internal structure is essential for the design of advanced devices exploiting nanoscale ferroicity and localized functional properties.

Second-harmonic generation (SHG) microscopy allows for the non-perturbative observation of ferroelectric domains and domain walls. The method can provide important insight into the structure of domain walls since the optical susceptibility tensor describing the SHG process is directly related to the local crystal symmetry and to the ferroic order.

We use SHG microscopy to probe the internal structure of 180° ferroelectric domain walls in lead titanate thin films and lithium tantalate bulk crystals. In both systems we detect a pronounced SHG signal at the walls. Local polarimetry analysis of this signal combined with numerical modeling reveals the existence of a planar polarization within the walls, with Néel and Bloch-like configurations in lead titanate and lithium tantalate, respectively. Depth-resolved SHG microscopy is employed to analyze the three-dimensional DW structure in the lithium tantalate bulk crystal. We thereby find that the chirality of Bloch-type walls may change at line defects, in close analogy to Bloch lines in ferromagnets.

Our results demonstrate a clear deviation from the ideal Ising-type domain wall configuration that is traditionally expected in uniaxial ferroelectrics. This corroborates recent theoretical predictions of a more complex, often chiral domain wall structure.