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VIEWPOINT

The many surprises of ferroelectric superlattices

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Ferroelectricity is a collective phenomenon: it involves the cooperative parallel alignment of electrical dipoles (and in the small print of definitions, there is also a requirement that the direction of dipole alignment can be reversed by application of an external electric field). Dipole interaction is therefore central in determining the properties of ferroelectric materials. Such interactions can be relatively long range—estimates for the critical correlation volume of cooperating dipoles below which ferroelectricity cannot exist vary, but are typically between 10 and 100 nm³. In addition, since electrical dipoles are created by the physical separation of ions, there is also a strong interaction between ferroelectricity and any strain in the crystal. Given these circumstances, it is obvious that in the creation of superlattice structures with periodicities capable of interrupting the critical correlation volume, using two different ferroelectric materials with different chemistry, dipole behaviour and lattice parameters, we may well expect to see radical alteration of material properties.

Experimentally, effects have indeed been found. The most commonly observed phenomenon is an enhancement in the dielectric constant as the superlattice periodicity is reduced. Typically fine-scale superlattices will demonstrate dielectric constants around twice those of coarse ones [1–4]; permittivities are also significantly greater than those possessed by single-layer solid-solution thin films, with the same global composition as that of the combined superlattice components [5]. Occasionally, outlandish claims have been made. For example, in 1996 Erbil *et al* [6] published a low-frequency dielectric constant of 420 000—three or four orders of magnitude greater than the permittivities typically seen in thin films. However, readers should always view such claims with healthy scepticism. Recent articles by O'Neill *et al* [7] and Catalan *et al* [8] have highlighted the likely role of defect concentrations at superlattice interfaces in causing apparent dielectric enhancement. The general problem of misinterpreting giant permittivities is also well discussed by Lunkenheimer *et al* [9], but it seems that it is a particular danger in the functional characterization of superlattice structures.

Elevated remanent polarization (polarization in the absence of an applied electric field) is also a feature of fine-scale superlattices, as demonstrated by Shimuta *et al* [10]. In their work, superlattices of BaTiO₃ and SrTiO₃, with three unit cells of each in the superlattice period, demonstrated a remanent polarization (P_r) greater than that of single-phase BaTiO₃ thin films.

So-called ‘asymmetric superlattices’ (with more BaTiO₃ than SrTiO₃ in the superlattice period) showed an even more dramatic response, with the maximum observed P_r more than three times that of single-phase BaTiO₃ thin films, and approaching that of bulk BaTiO₃.

Scientifically, both elevated dielectric constant and remanent polarization are extremely interesting, but they also have technological relevance—both parameters are critical in a variety of thin-film memory applications [11, 12]. Importantly though, a detailed understanding of the origins of the unusual properties displayed by ferroelectric superlattices is not yet evident. To get to the heart of the matter, insight into the development of polarization, and polar dynamics in superlattice structures is required. Occasional glimpses can be found by electrical testing—for example the observation of antiferroelectric coupling in KNbO₃/KTaO₃ systems [13]. However, on the whole, direct electrical characterization is fraught with danger, as there is great potential for interpreting the effects of defects as fundamental rather than peripheral. In general, insight gained by structural or crystallographic investigation is more robust [14, 15].

In issue 21 of *Journal of Physics: Condensed Matter* Ríos *et al* [16] report on an x-ray diffraction study on SrTiO₃–BaTiO₃ superlattices performed at room temperature. Within the superlattice structure, the BaTiO₃ was found to be tetragonal, as would be expected, with the primary tetrad axis oriented parallel to the superlattice periodicity (i.e. out of the plane). This is the axis along which the electric dipole or polarization in tetragonal BaTiO₃ exists. On the other hand, the symmetry of the SrTiO₃ was observed to be orthorhombic, in stark contrast with its expected room temperature cubic symmetry. For orthorhombic SrTiO₃, polarization occurs along the pseudocubic [110] and [1 $\bar{1}$ 0] directions. The implication is that the polarization in the SrTiO₃ layers is at 90° to that in the BaTiO₃—a hypothesis well supported by second-harmonic generation experiments performed on the same system by Jiang *et al* [17]. Essentially the interfaces between SrTiO₃ and BaTiO₃ are analogous to 90° domain walls. The work represents a clean and uncluttered view of the symmetry and hence implied polarization states in the superlattice structure. It is *new* insight and, while it is not immediately obvious how this relates to the surprising functional properties of superlattices described above, it is surely an important early step in our understanding.

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