

Computing Spontaneous Polarization Without Sampling a Switching Path

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The spontaneous polarization of a ferroelectric is a characterization of the current which flows when the system is switched between two states. Computation of this spontaneous polarization in crystal systems has been enabled by the modern theory of polarization which relates the spontaneous polarization to a change in Berry Phase as the material switches. Computing this phase change modulo 2π is quite straightforward. The measured spontaneous polarization depends on the actual path along which the material switches, which in general involves nucleation and growth of domains and is therefore quite complex. Existing schemes to predict this measured value from first principles involve dividing the total phase change into smaller changes by computing the total phase at intermediate points along a particular (typically uniform) fictitious switching path. This can be problematic both due to the additional computational resources required and challenges in automatically identifying a physically relevant path which keeps the system insulating. In this work we present a method of predicting the spontaneous polarization which involves separating the change in phase between two states into as many gauge invariant smaller phase changes as possible. As long as the magnitude of these smaller phase changes remain much smaller than π , their sum forms a phase change which corresponds to the change one would find along any path involving minimal evolution of the atomic and electronic structure. We show that for many ferroelectrics, including those which would have otherwise required a densely sampled path, this technique allows for the spontaneous polarization to be computed without any need for intermediate sampling between oppositely polarized states at all.