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Supplementary Materials for

Theoretical guidelines to create and tune electric skyrmion bubbles

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This PDF file includes:

Supplementary Discussion I: Terminology considerations

Let us comment on a recurrent terminology issue, namely, whether objects as those discussed in this work should be termed "skyrmions" or "chiral bubbles". As far as we understand from the magnetic literature, both names denote the same kind of objects when considered individually, and the difference seems to be related to the collective behavior of a lattice of skyrmions or bubbles: chiral bubbles would be weakly coupled to each other, with frequent helicity changes from one bubble to the next, yielding no macroscopic chirality; in contrast, skyrmions would occur in ordered lattices displaying a long-range chiral order. Strictly speaking, the present work does not address the behavior of the "skyrmion/bubble lattice" and, thus, we do not have any argument to favor one terminology over the other when referring to the objects here investigated. Hence, in this article we use the term "skyrmion bubble" as a compromise that hopefully will not be misleading.

The obtained electric skyrmion bubble (ESB) is obviously chiral, with a non-zero helicity that remains constant as we move along z. Our ESB is strongly reminiscent of the so-called *magnetic soft bubbles* discussed in the literature on magnetic skyrmions (see e.g. Fig. 2.6d of Ref. 1); more specifically, ours would be a Bloch-type bubble. In addition, we can of course use a spherical representation of the electric dipoles in our skyrmion bubble, as shown in Suppl. Fig. 1; in this way, the correspondence with the type of *hedgehog skyrmions* sometimes called *combed* becomes apparent [36].

Supplementary Discussion II: Minimal size of ESBs

One may wonder: how small may our electric skyrmion bubbles get to be? The questions is particularly interesting because our ferroelectrics does not present any analogue of the magnetic Dzyaloshinskii-Moriya interactions (DMI), while the smallest magnetic skyrmions occur thanks to DMI couplings. Hence, can we have ESBs presenting a point-like core and a continuous rotation of the local dipoles, as the smallest magnetic skyrmions do? To address these issues, let us make the following points.

According to our simulations of $PbTiO_3$, these ESBs will appear whenever we write column domains in a matrix of opposed polarization. Since the column domain constitutes the core of the skyrmion, the question about the minimum skyrmion size amounts to asking: how small can a column domain be? Let us further stress that we assume we can write the column domains; discussing the smallest possible size of written domains is very different from addressing the preferred size of spontaneously-occurring skyrmions/bubbles.

Now, let us explan our way to think about this minimum size. From first-principles simulations [37], we know that a single column of parallel-aligned electric dipoles, with a section of one 5-atom unit cell, constitutes an instability of the cubic phase of many perovskite oxides. Further, we find it plausible that such a thinnest dipole column be stable within a matrix of opposed polarization. Note that this is not so different from the kind of structures occurring in antiferroelectrics, and that we know (again from first principles) that many ferroelectric materials present competing antiferroelectric phases [38]. Hence there is no fundamental reason against having electric skyrmions with very small cores, despite the absence of DMI-like interactions. In fact, this behavior may be relatively easy to obtain in perovskite oxides whose cubic phase have a very flat band of unstable polar phonons – as it is the case of $BaTiO_3$ [37] or $PbZrO_3$ [38].

To make this discussion more specific, we used our second-principles model for $PbTiO_3$ to consider the smallest possible column domain that is dynamically stable. Supplementary Figure S3 shows the result obtained when we consider a column domain of 1×1 unit cells in the plane: we have a stable skyrmion with a point-like core! Hence, even in $PbTiO_3$, which is not ideal material for these purposes as it features a relatively dispersive band of polar phonons, we are able to stabilize the smallest conceivable electric skyrmion!

Admittedly, this result is obtained in the limit of very low temperatures (strictly speaking at 0 K), and one should check the stability of such tiny skyrmions upon heating. Yet, let us stress that the Monte Carlo simulations described in this work indicate that skyrmions with a diameter of about 6 unit cells are stable up to room temperature, in spite of the fact that the boundary conditions assumed in the simulations (short circuit) do not favor their stability.

Now, do such tiny electric skyrmions present a "continuously rotating polarization"? We think the answer to that question may be in the eye of the observer. When our ESBs are very small, the core and the "rotation region" (i.e., the domain wall) become comparable in size. This automatically implies that we have large polarization gradients, i.e., rather abrupt changes of polarization that one would hardly describe as "continuous". At the same time, the in-plane component of the polarization in Suppl. Fig. 3 might be described as continuously rotating as, indeed, the associated rotational is non-zero everywhere in the domain wall. Hence, we are not really sure how to answer the above question. Nevertheless, as a matter of principle, it seems clear that the absence of DMI-like interactions does not preclude the occurrence of electric skyrmion bubbles bearing obvious similarities with the smallest magnetic skyrmions.

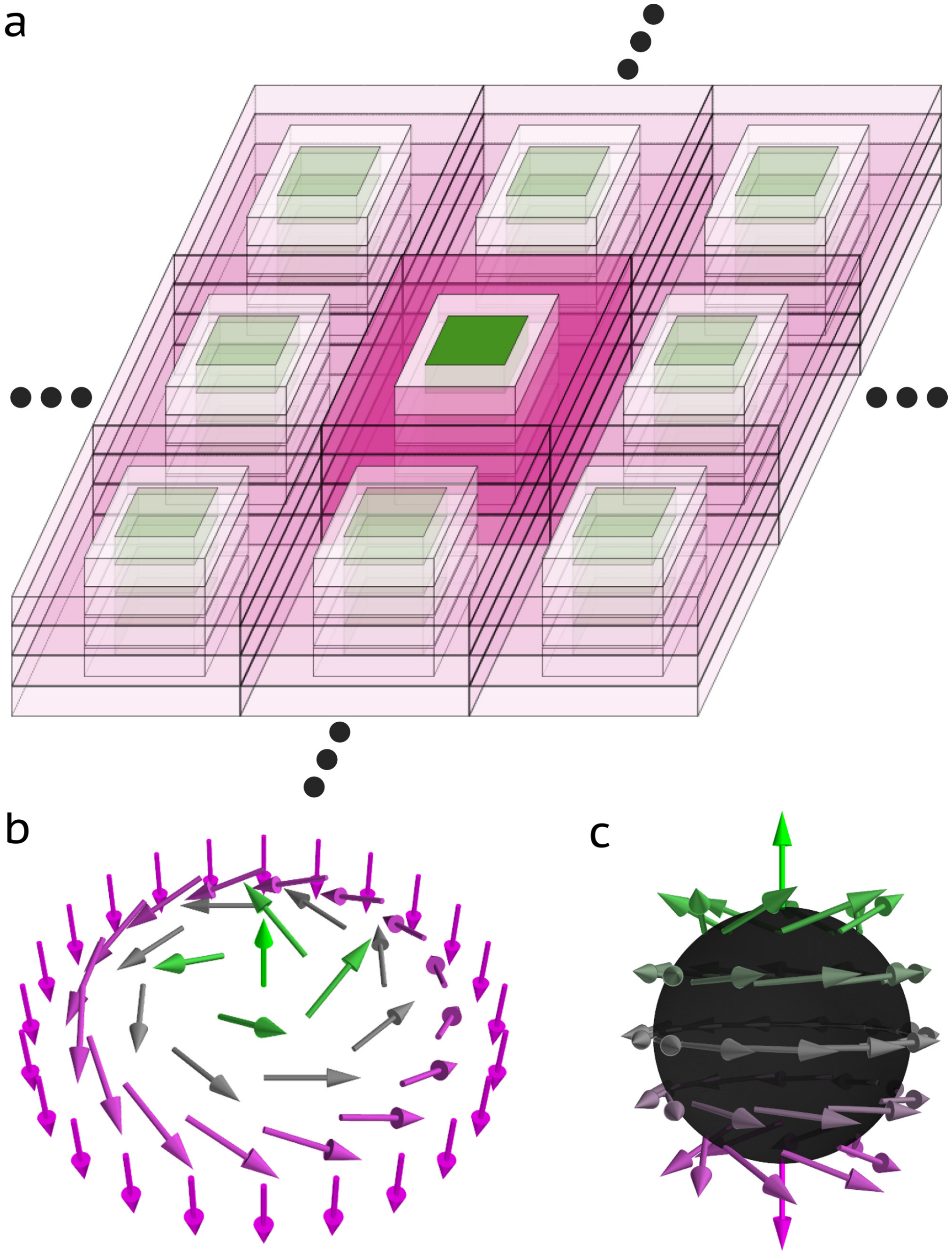

Fig. S1. **Sketches of simulation supercell and skyrmion structures.** Panel (a): Array of periodically-repeated NDs as considered in our simulations. Panel (b): Chiral skyrmion in a plane, analogous to the one discussed in this work. Panel (c) Spherical representation of the chiral skyrmion of panel (b).

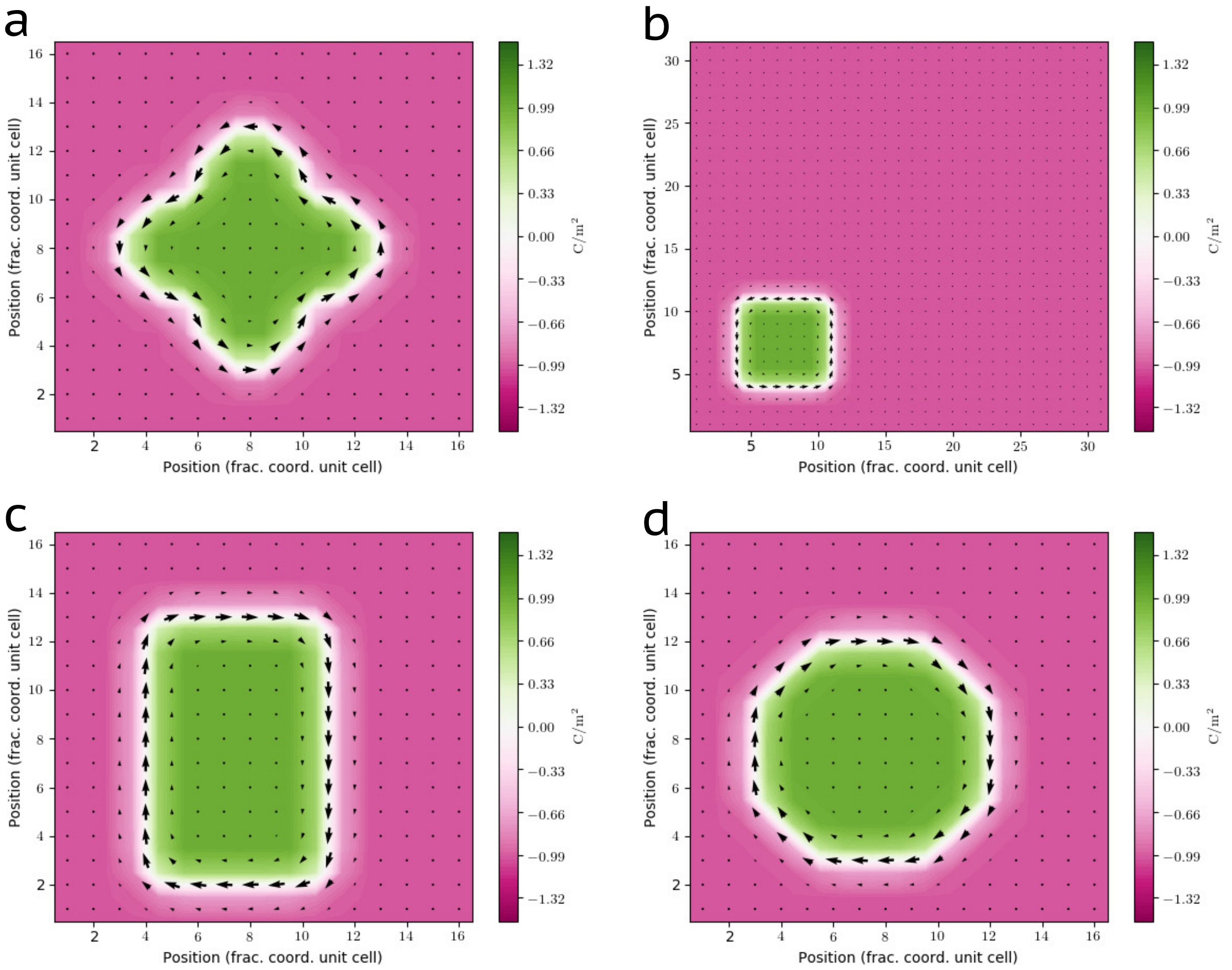

Fig. S2. **ESBs of different sizes and shapes.** Polarization maps obtained for different shapes and sizes of the ND. Panel (a): ND with NDWs in {110} planes. Panel (b): 6×6 ND within big 32×32 supercell (big matrix). Panel (c): rectangular 6×10 ND. Panel (d): Circular ND with a radius of about four perovskite cells. In panels (a) and (d) it is apparent that the relaxation changes the shape of the ND, as our model predicts a preference for the DWs to lie within {100} and {010} planes.

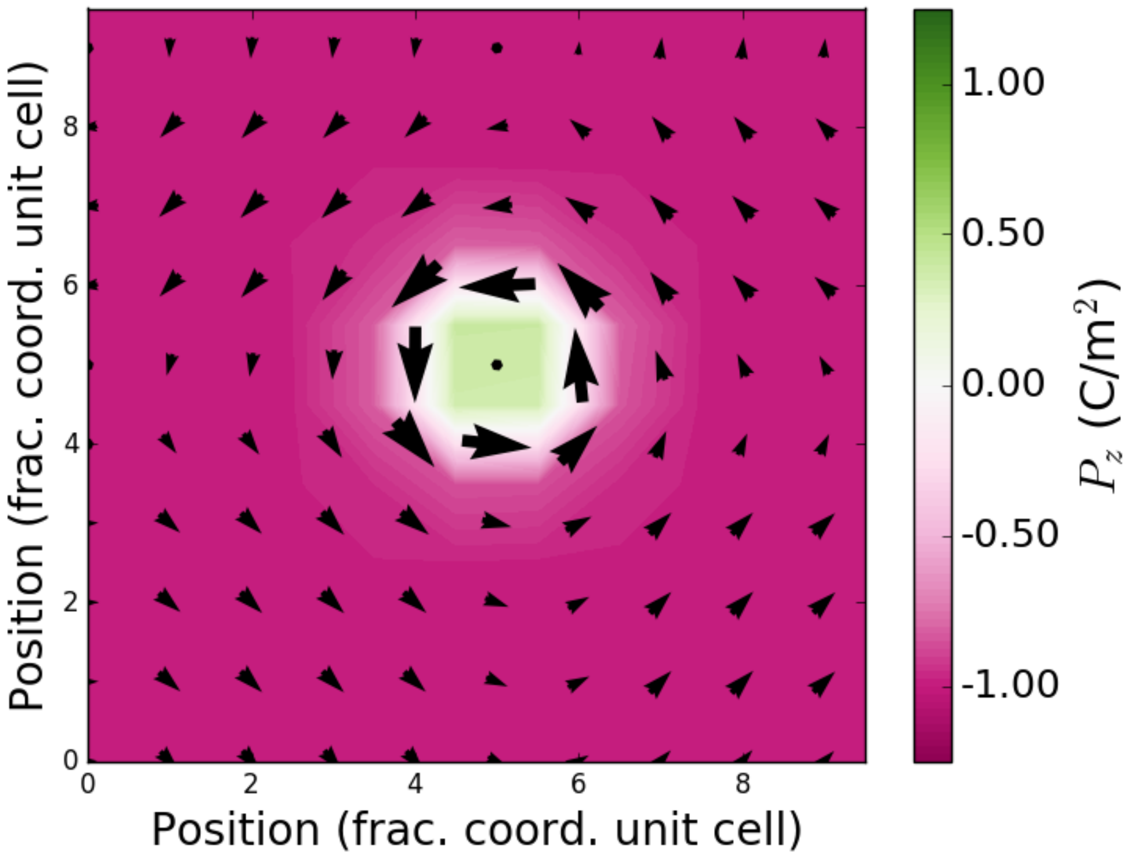

Fig. S3. **Smallest ESB.** Structure of the smallest imaginable electric skyrmion bubble, with a core of 1×1 unit cells, as obtained from our second-principles simulations by providing a suitable initial configuration and running a simulated-annealing relaxation.

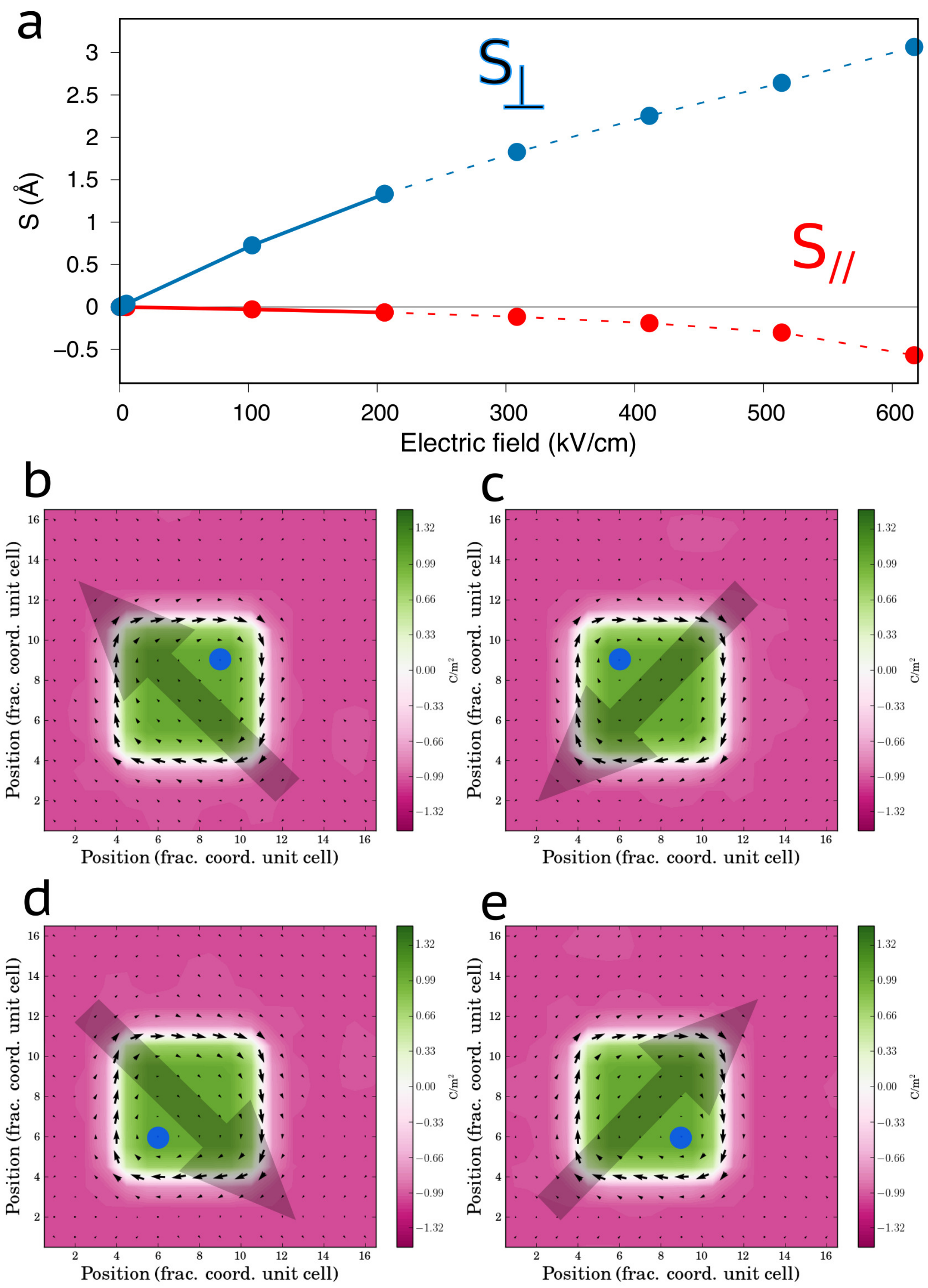

Fig. S4. **ES**B **as a four-state memory.** Panel (a) Drift of the skyrmion center as a function of an in-plane electric field along (,11); we show the results along the directions parallel (red) and normal (blue) to the field. The dashed line marks the range where the NDW-polar state becomes the ground state and the polar-ESB state is metastable. Panels (b–e): Polarization maps obtained when an in-plane electric field (indicated by the shadowed arrow) is applied to the polar-ESB state. The field can be used to switch the position of the ESB center, as emphasized by the blue circles.