

The Monte Carlo simulation models a layered AFM insulator with four anti-ferromagnetically coupled layers, each composed of 40×40 spins. A single-spin Metropolis algorithm is used and energy fluctuations are determined by the system Hamiltonian,

$$H = -J_{FM} \sum_l \sum_{\langle i,j \rangle} \vec{S}_{l,i} \cdot \vec{S}_{l,j} + \sum_{l < N} J_{AFM,l} \sum_i \vec{S}_{l,i} \cdot \vec{S}_{l+1,i} + \sum_{l,i} K_l (S_{l,i,z})^2 - B \sum_{l,i} S_{l,i,z}$$

where $\vec{S}_{l,i}$ is the spin unit vector residing on site i of layer l . The J_{FM} term characterizes intra-layer ferromagnetic interactions and the $J_{AFM,l}$ term characterizes antiferromagnetic coupling between layers l and $l + 1$. The K_l term describes easy-axis anisotropy (for $K_l < 0$) and the B term describes Zeeman coupling. In our simulations, $J_{FM} = 1$ and magnetization per spin is calculated by averaging the $S_{i,z}$ components of all spins. When sweeping the magnetic field strength, magnetization per spin is measured over the entire system as well as on each individual layer. Mean magnetization per spin measurements are calculated by averaging the mean $S_{i,z}$ value over 1000 ensembles at the indicated external field strength.

As mentioned in the main text, applying a gate voltage to the top or bottom of the system breaks inversion symmetry. This effect is modeled in our simulation by adjusting the anisotropy K or interlayer coupling strength J_{AFM} on the top and bottom layers. When sweeping the magnetic field strength in a system that exhibits inversion symmetry (i.e. uniform parameters throughout every layer), we observe all of the patterns of Figure 1 with equal probability.

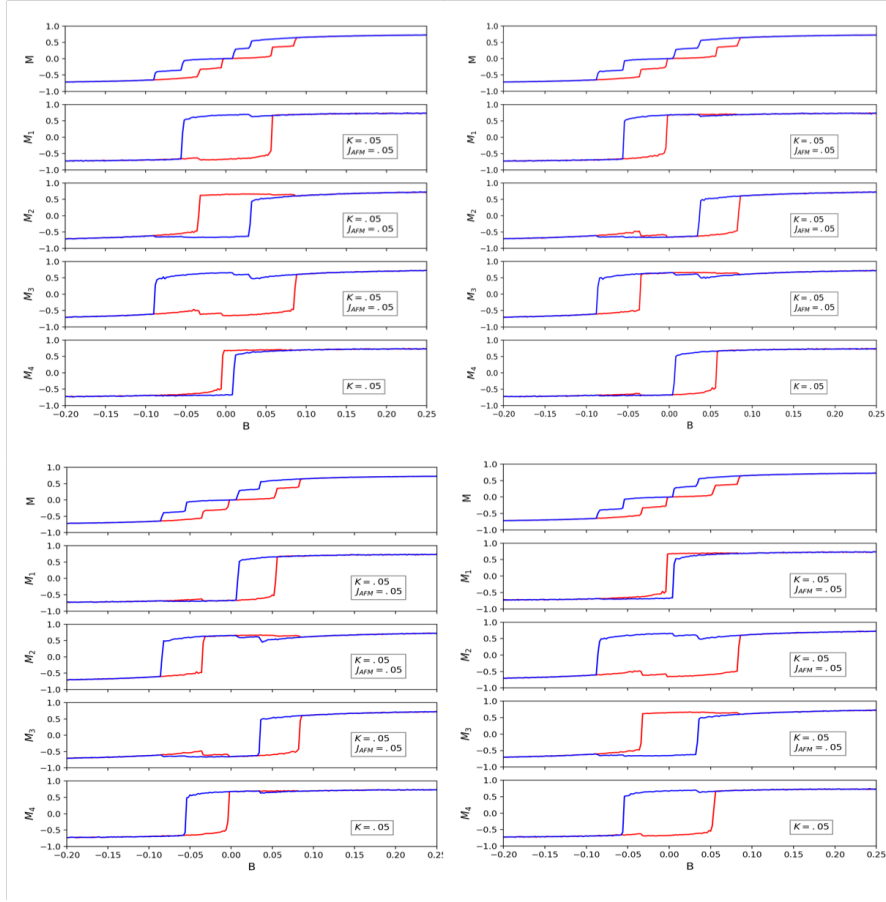


Figure 1: Magnetization versus magnetic field strength B for the total system, M , and each of the individual layers, M_i . Shown are the four possible switching patterns obtained for a symmetric system when sweeping the magnetic field. The red line indicates sweeping of the magnetic field strength in the positive direction, and the blue line indicates sweeping in the negative direction. All results presented are for four square lattice layers of 40×40 spins each at $T = .15$. The anisotropy strength K and the antiferromagnetic coupling J_{AFM} are shown in the inset of the individual layer plots, where J_{AFM} displayed on the plot for layer i refers to the coupling between layers i and $i + 1$.

One possible approach for breaking inversion symmetry is to modify the anisotropy K on a single layer. An example of this approach, where K_1 is increased from .05 to .08, is shown in Figure 2. In increasing the anisotropy on the top layer, we observe behavior similar to that shown in Figure 2(b) in the main text. That is, when reversing the sweeping direction, the preferred bistable state in the intermediate magnetic field region is switched. The observed states

for this case are enumerated in Figure 3. We can obtain similar behavior by increasing J_{AFM} between the top two layers. An example of this scenario is shown in Figure 4, where J_{AFM} is increased from .05 to .08 between the top two layers only.

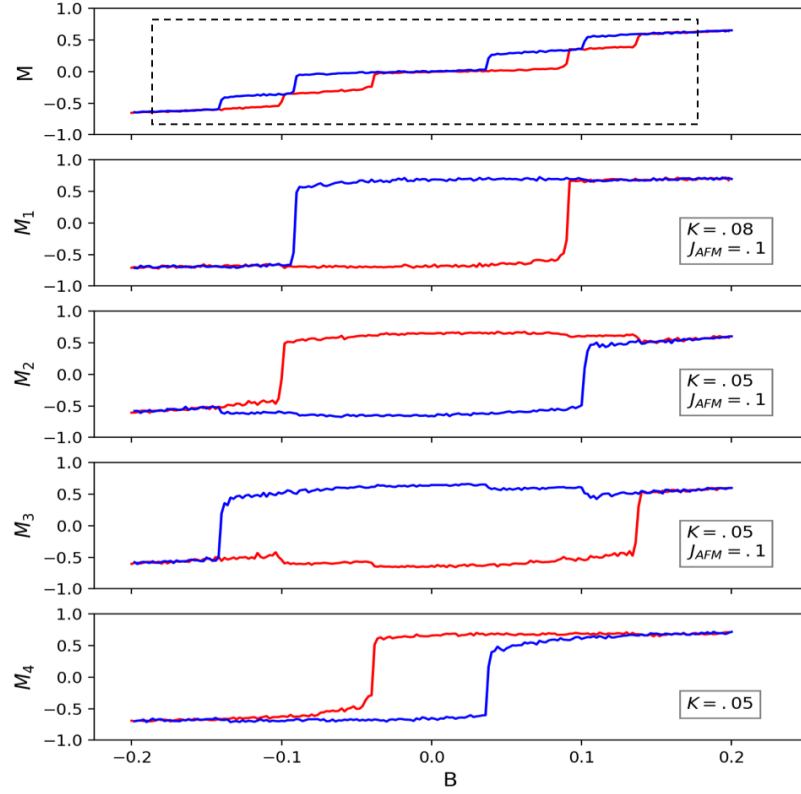


Figure 2: The anisotropy of the top layer is increased slightly to $K_1 = .08$ while the other layers have an anisotropy of $K = .05$. The values of J_{FM} and J_{AFM} are uniform throughout the system. The states observed in each intermediate field region within the dashed lines are shown in Figure 3.

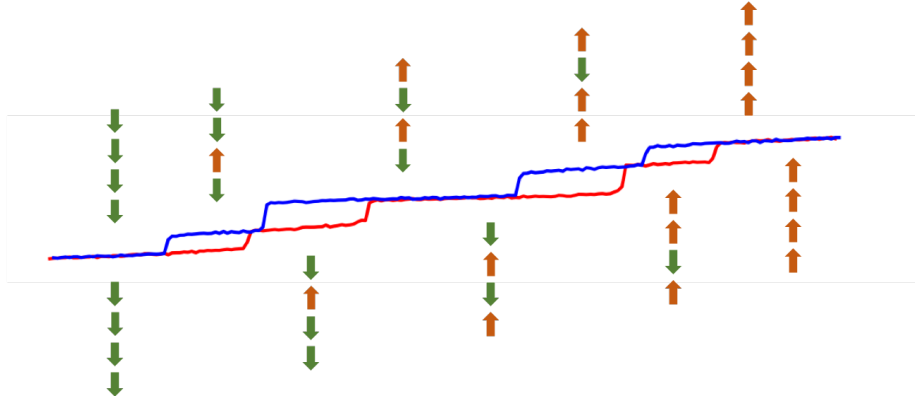


Figure 3: The preferred states observed in the dashed line region of Figure 2. States observed during the positive sweep are shown below the plot, and states observed during the negative sweep are shown above. In the positive intermediate field region, for example, we see that $\uparrow\downarrow\uparrow\uparrow$ is preferred during a negative sweep while $\uparrow\uparrow\downarrow\uparrow$ is preferred during a positive sweep.

If we increase the interlayer coupling strength in addition to breaking the symmetry of the system, we begin to see a shift from the behavior described in Figure 2(b) of the main text to that of Figure 2(a) or 2(c). That is, the preferred bistable state in a subset of the intermediate magnetic field region will no longer depend on the sweeping direction. As an example, if we increase J_{AFM} to .3 between the top two layers (Figure 5), the results of sweeping the magnetic field become similar to those obtained in Figure 2(c) of the main text. In particular, we observe that $\uparrow\downarrow\uparrow\uparrow$ is preferred in a small positive intermediate field region during both the positive and negative sweep of the magnetic field. Another example of this behavior is also shown in Figure 5 for an increased anisotropy on the top layer.

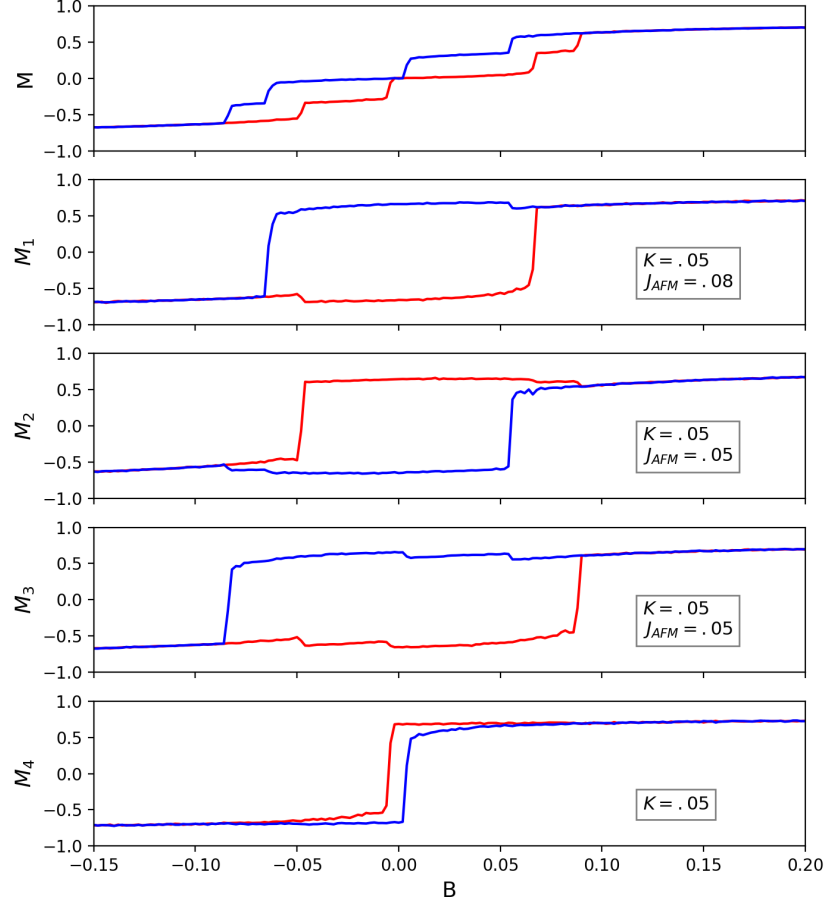


Figure 4: The interlayer coupling of the top two layers is increased to $J_{AFM} = .08$ while the other two interlayer couplings are kept at $J_{AFM} = .05$. The values of K and J_{AFM} are uniform throughout the system.

We now turn to a simulated demonstration of controlled switching between bistable states. Consider the results presented in Figure 2. The bistable states present in the positive intermediate field region are highlighted in Figure 6. We see that the $\uparrow\uparrow\downarrow\uparrow$ state is preferred during the positive sweep of the magnetic field, while $\uparrow\downarrow\uparrow\uparrow$ is preferred during the negative sweep. We also note that the intermediate field region where the bistable states are encountered is bounded

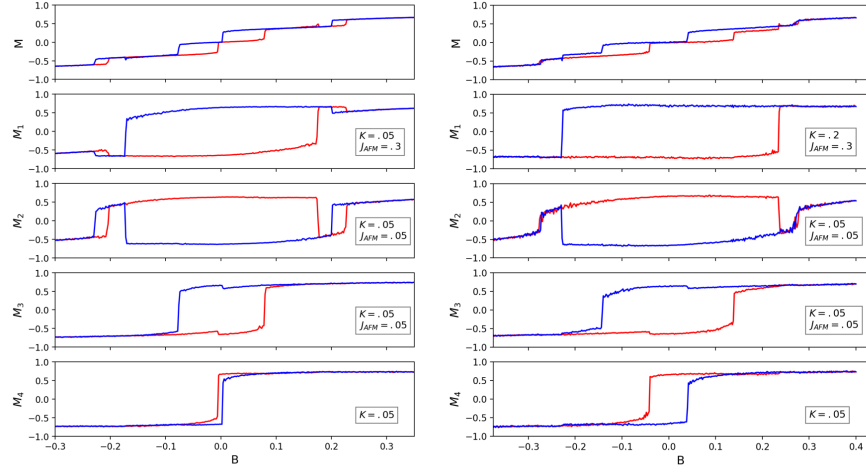


Figure 5: In this simulation, the interlayer coupling strength of the top two layers is increased significantly to $J_{AFM,12} = .3$ while the other layers have a coupling strength of $J_{AFM} = .05$. The values of On the left, J_{FM} and K are uniform throughout the system. On the right, the anisotropy K is also increased on the top layer.

by two critical field values B_0 and B'_0 . By initially sweeping the magnetic field from some large positive value (where $\uparrow\uparrow\uparrow$ is guaranteed) to $B_0 + \Delta$, we can obtain the initial bistable state $\uparrow\downarrow\uparrow$. If Δ is small, we can then fix the magnetic field strength at $B_0 + \Delta$, which is very near the critical field value B_0 . By then sweeping the overall anisotropy strength, we can shift the critical switching field, moving the system temporarily out of the intermediate field region. As we approach the critical field from the positive sweeping direction, the preferred bistable state switches to $\uparrow\uparrow\downarrow$. This process is demonstrated in Figure 7.

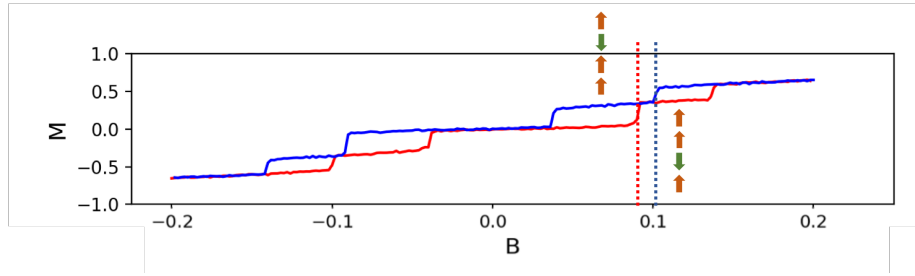


Figure 6: We see that the $\uparrow\uparrow\downarrow\uparrow$ state is preferred during the positive sweep of the magnetic field, while $\uparrow\downarrow\uparrow\uparrow$ is preferred during the negative sweep. The critical field values B_0 and B'_0 that bound the positive intermediate field region where these states are present are indicated by red and blue dashed lines, respectively.

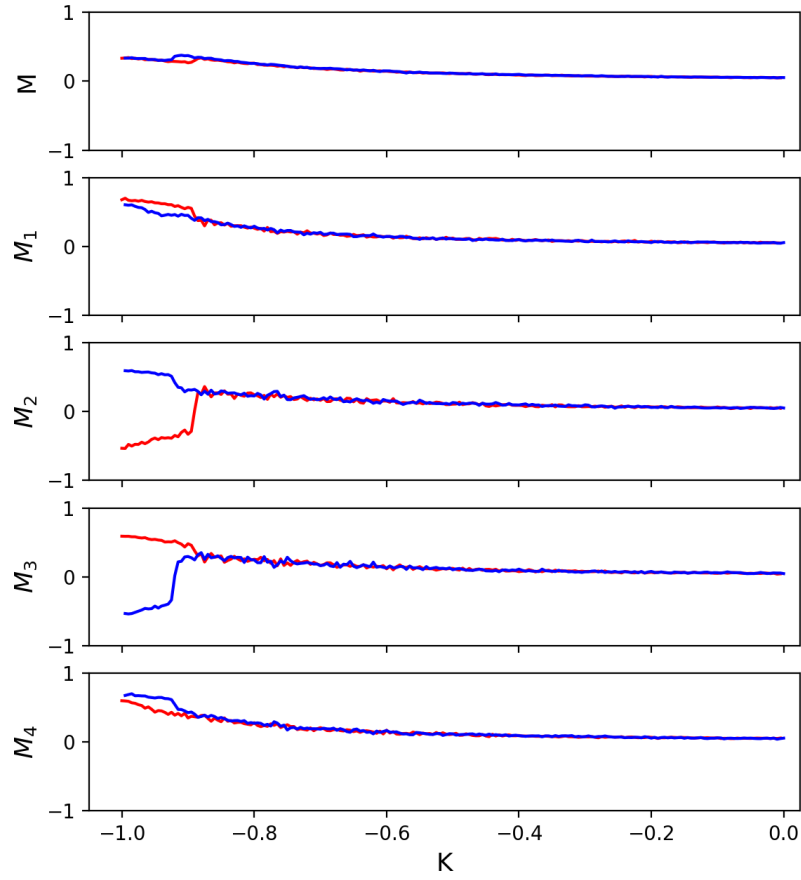


Figure 7: By sweeping the overall anisotropy, we are able to perform a controlled switch between bistable states as the magnetic field is fixed at $B = .09$. The system is initially in the state $\uparrow\downarrow\uparrow\uparrow$, but after sweeping the anisotropy, we obtain the state $\uparrow\uparrow\downarrow\uparrow$.