Hybrid improper ferroelectricity and magnetoelectric coupling in a two-dimensional perovskite oxide

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We demonstrate that the ferroelectricity induced by octahedral rotation and the magnetoelectric coupling mechanism dependent on Dzyaloshinskii-Moriya (DM) interaction exist in a proposed perovskite bilayer. Although the octahedral rotation distortion of the $Ca_3Mn_2O_7$ bilayer is reconstructed with respect to its layered bulk phase, tensile strain can induce the same octahedral rotation type as its bulk phase, resulting in the emergence of ferroelectric phase. We show that the modulation of the DM vector by octahedral rotation distortion provides a coupling between polarization and the induced magnetization. In ferroelectric switching, the polarization is reversed by a 180° rotation of the tilt axis within the ab plane, which also results in a reversal of magnetization. This work is expected to provide a guidance for realizing electric-field control of magnetization direction in 2D perovskite oxides.

➢ Ground-state structure

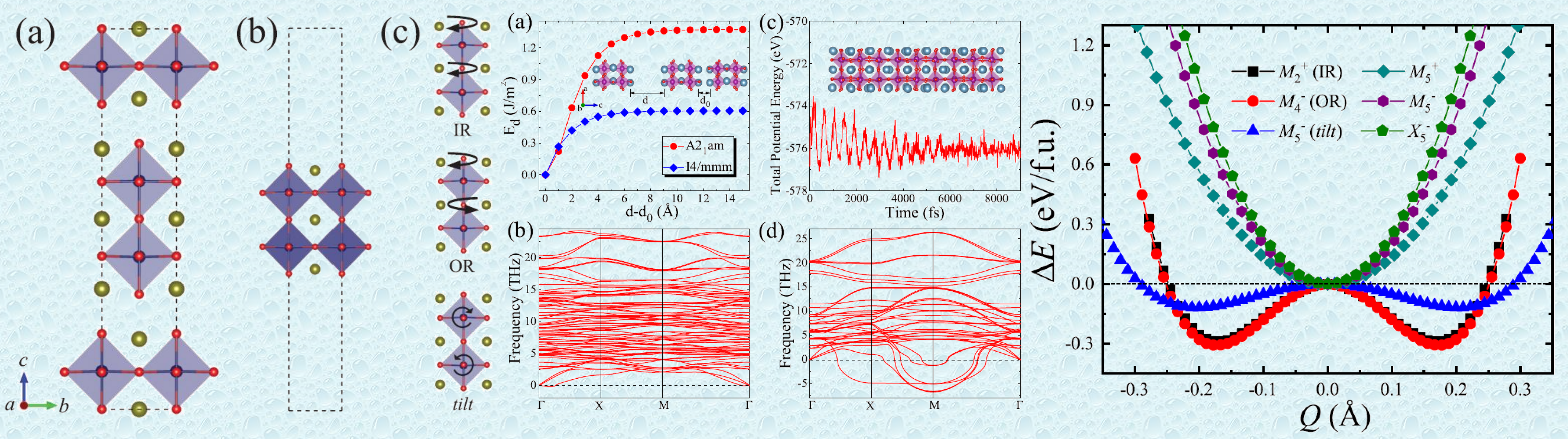
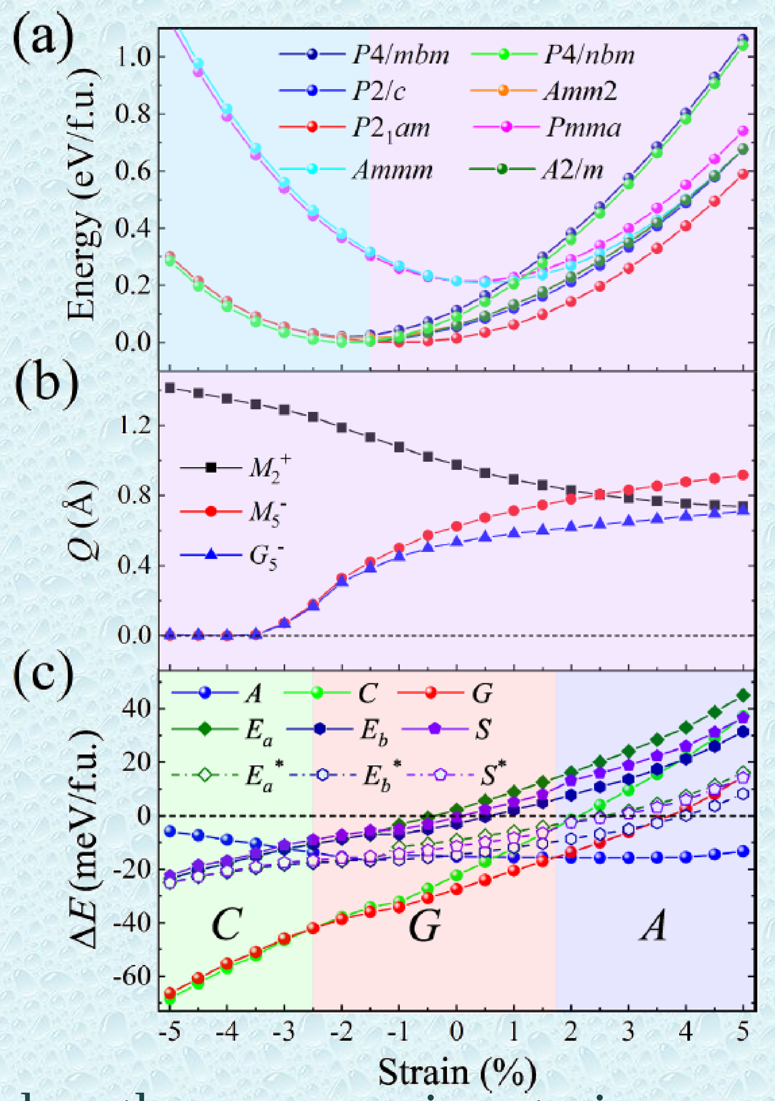


TABLE II. Symmetry and energy of structural phases resulting from various octahedral rotation types for the strain-free $Ca_3Mn_2O_7$ bilayer.

Irrep	Space group	ΔE (meV/f.u.)
$M_2^+(a)$	*P*4/*mbm* (No.127)	20.6
$M_4^-(a)$	*P*4/*nbm* (No.125)	0
$M_5^-(a;0)$	*Pmma* (No.51)	210.4
$M_5^-(a;a)$	*Ammm* (No.65)	224.2
$M_5^-(a;b)$	*P*2/*m* (No.10)	–
$M_2^+(a) \oplus M_5^-(b;0)$	*P*2$_1$*am* (No.26)	5.6
$M_2^+(a) \oplus M_5^-(b;b)$	*Amm*2 (No.38)	16.4
$M_2^+(a) \oplus M_5^-(b;c)$	*Pm* (No.6)	–
$M_4^-(a) \oplus M_5^-(b;0)$	*P*2/*c* (No.13)	0.2
$M_4^-(a) \oplus M_5^-(b;b)$	*A*2/*m* (No.12)	0.1
$M_4^-(a) \oplus M_5^-(b;c)$	$P\bar{1}$ (No.2)	–

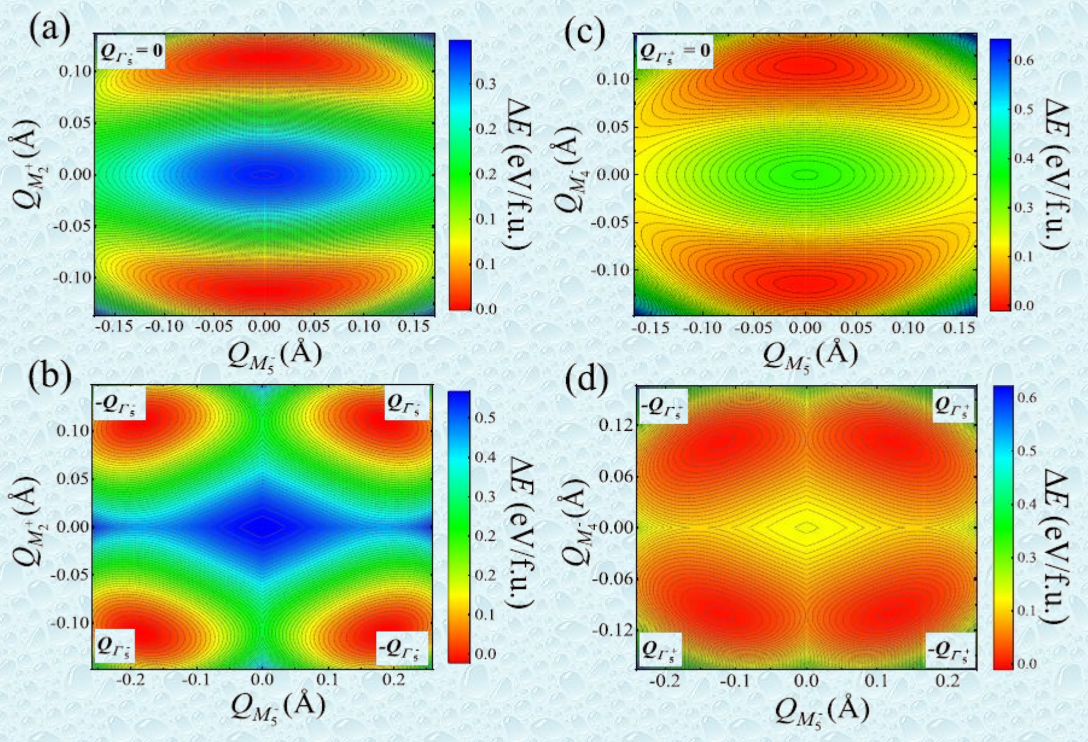
Table II shows that the nonpolar *P*4/*nbm* phase caused by $a^0a^0c^-$ type octahedral rotation has the lowest energy, that is, only the OR mode occurs in the ground-state structure. This means that the octahedral rotation type is reconstructed with respect to its bulk phase.

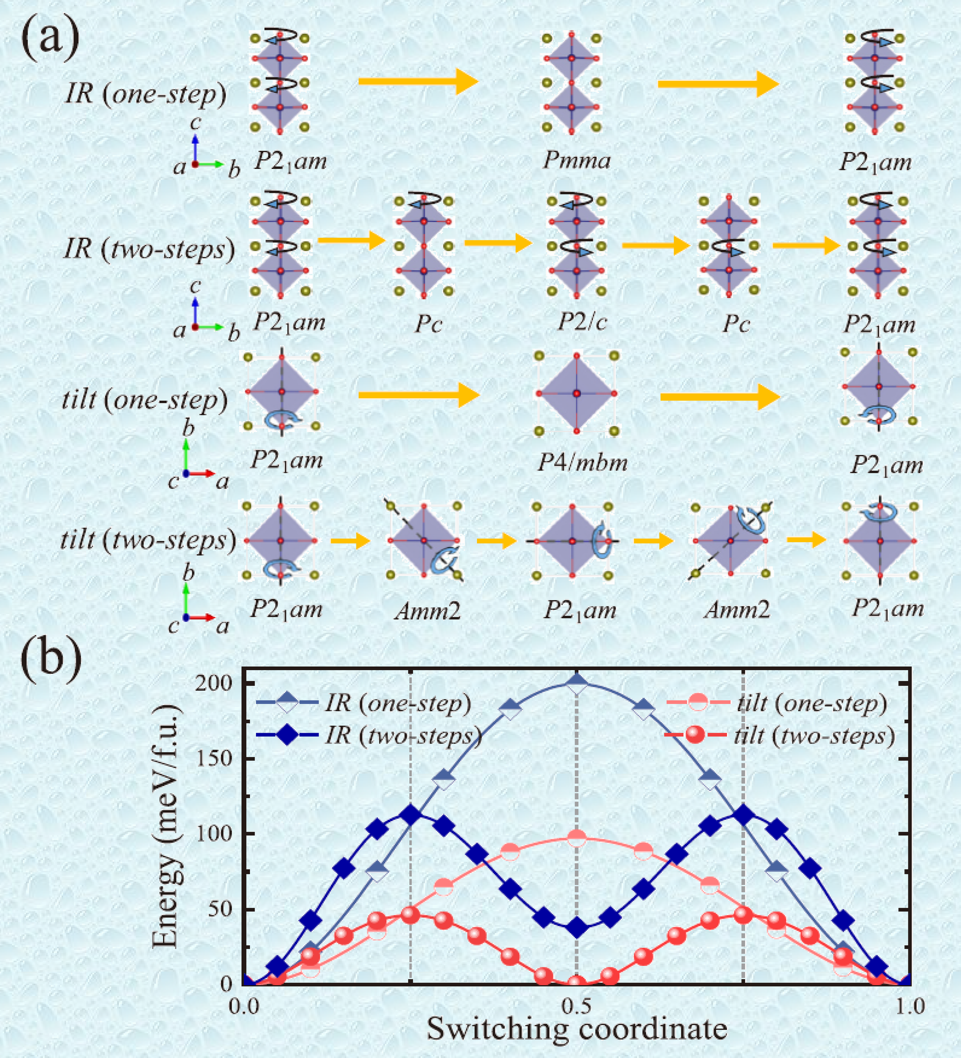
➢ Strain effect



when the compressive strain exceeds 1.5%, the nonpolar *P*4/*nbm* phase is the ground-state phase, however, the tensile strain causes the ground-state phase to transform into to the polar $P2_1am$ phase.

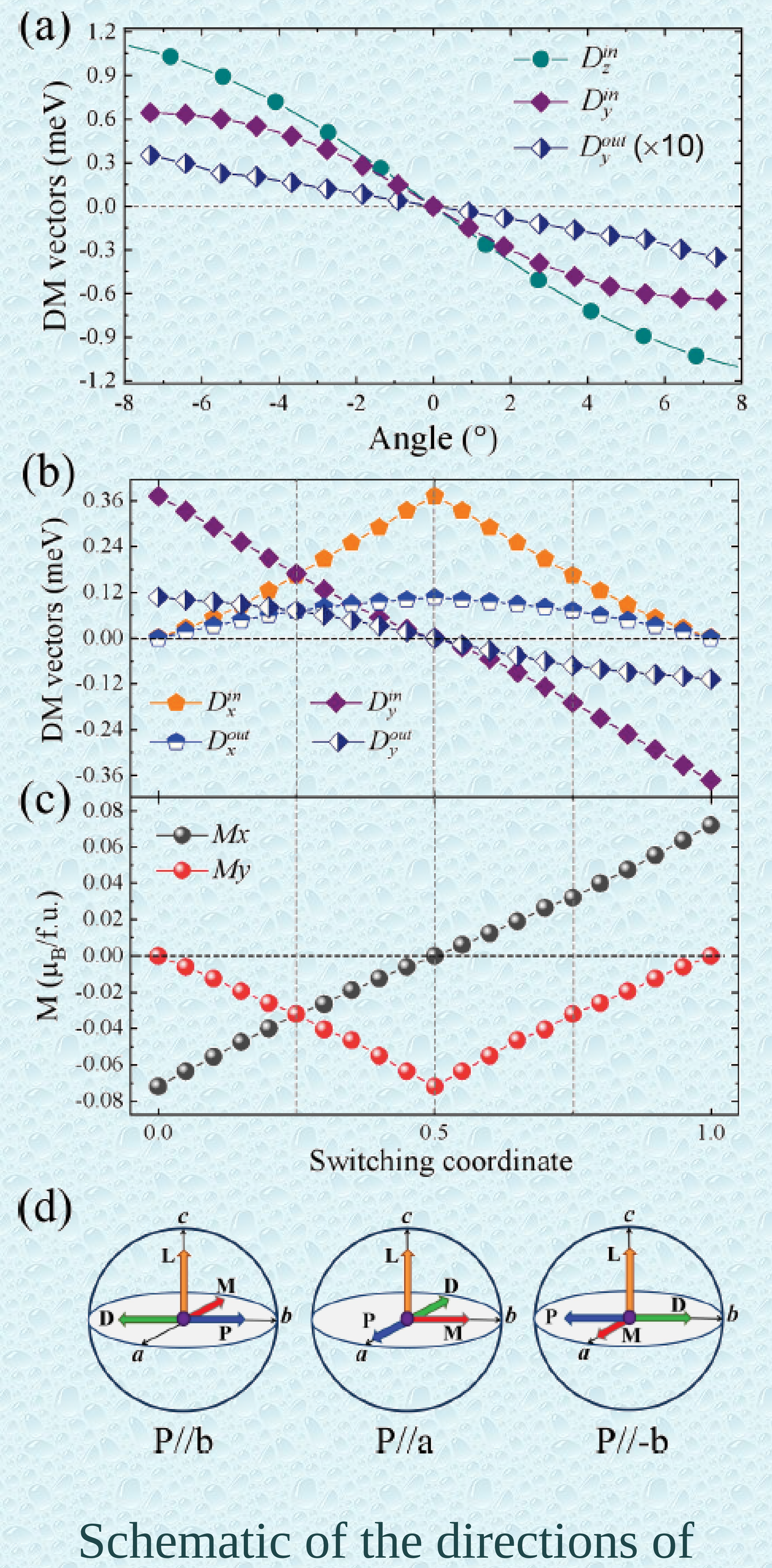
➢ Ferroelectric switching





The calculated switching barrier of the $Ca_3Mn_2O_7$ bilayer at 0% strain reaches 45 meV/f.u., which is much less than the calculated value (82 meV/f.u.) of the $Ca_3Ti_2O_7$ bulk.

➢ ME coupling



Schematic of the directions of polarization (**P**), AFM vector (**L**), DM vector (**D**), and the induced magnetization (**M**) in the initial, intermediate, and final states of the lowest-energy ferroelectric switching.

➢ Conclusion

In conclusion, we demonstrate the octahedral rotation induced improper ferroelectricity and ME coupling effect in a perovskite bilayer.

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