

Supplementary Information for "Ultrafast optical route to coupled ferroelectric and altermagnetic switching"

Yuhao Gu,^{1,*} Yu-hui Song,^{2,3,*} Peng-Jie Guo,^{2,3,*,†} Yihao Wang,¹ Zhe Li,¹ Ze-Feng Gao,^{2,3} Huan-Cheng Yang,^{2,3} and Zhong-Yi Lu^{2,3,4,‡}

¹*School of Mathematics and Physics, University of Science and Technology Beijing, Beijing 100083, China*

²*School of Physics and Beijing Key Laboratory of Opto-electronic Functional Materials & Micro-nano Devices, Renmin University of China, Beijing 100872, China*

³*Key Laboratory of Quantum State Construction and Manipulation (Ministry of Education), Renmin University of China, Beijing 100872, China*

⁴*Hefei National Laboratory, Hefei 230088, China*

* These authors contributed equally to this work.

† guopengjie@ruc.edu.cn

‡ zlu@ruc.edu.cn

I. MAGNETIC ORDERS

Here, we show the magnetic orders considered in our paper.

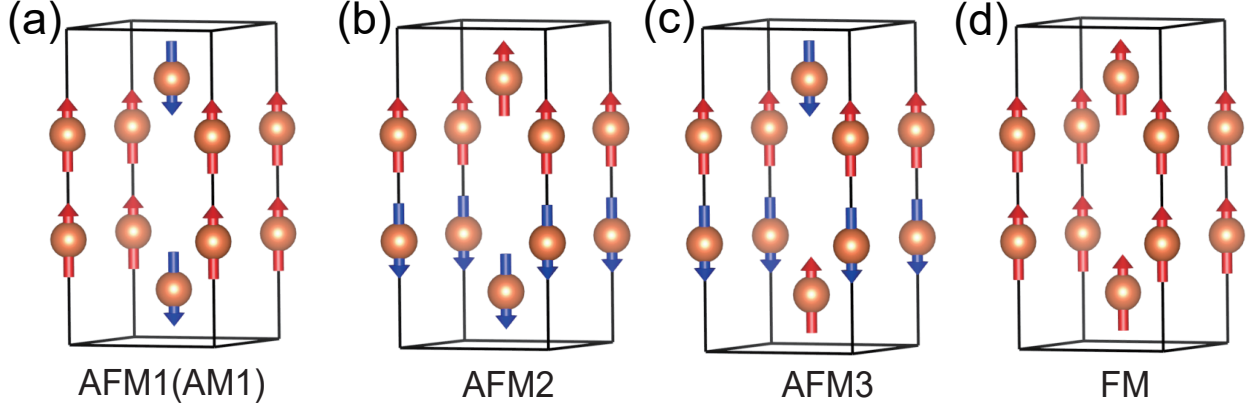


FIG. S1. The magnetic orders of HSS LiV_2F_6 considered in this paper. The orange balls represent $\text{V}^{2.5+}$ ions. Nonmagnetic Li and F atoms are hidden for simplicity.

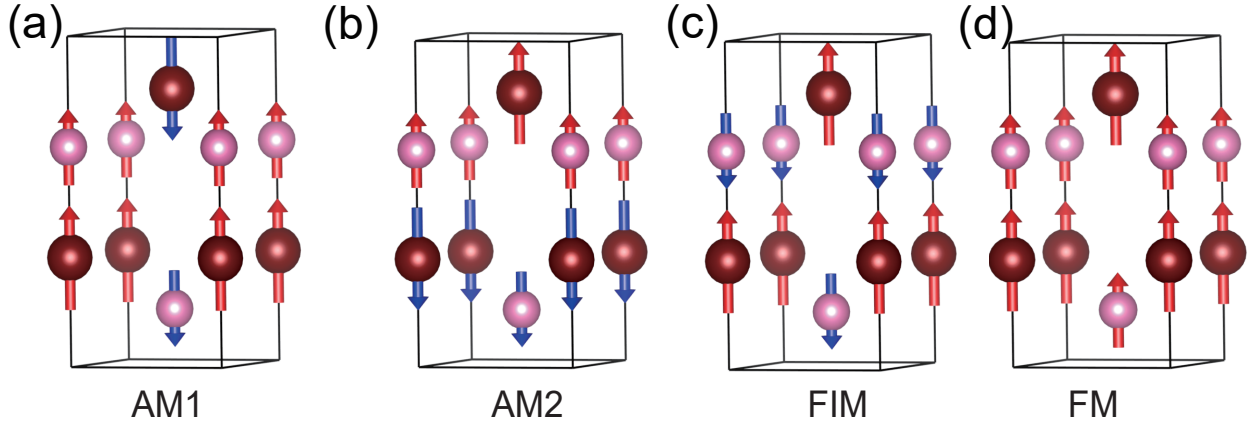


FIG. S2. The magnetic orders of LSS LiV_2F_6 considered in this paper. The dark red and pink balls represent V^{2+} and V^{3+} ions, respectively. Nonmagnetic Li and F atoms are hidden for simplicity.

II. DYNAMICAL STABILITY

To examine the structural stability of the proposed charge-ordered phase, we calculated the phonon spectra of both the paraelectric high-symmetry structure (HSS) and the charge-ordered ferroelectric low-symmetry structure (LSS), as shown in Fig. S3. Because the magnetic ground

state of the HSS structure is FM and the intermediate state in the switching process is AFM2, we calculated the phonon spectra of the HSS structure with these two magnetic orders. Both phonon spectra exhibit unstable phonon modes, indicating that the paraelectric structure is dynamically unstable against a symmetry-lowering distortion. In both cases, the largest imaginary-frequency mode appears at the Γ point ($q = 0$), suggesting that charge ordering may emerge without enlarging the crystallographic unit cell. In contrast, the LSS phase shows no imaginary phonon frequencies throughout the Brillouin zone, confirming the dynamical stability of the charge-ordered ferroelectric phase.

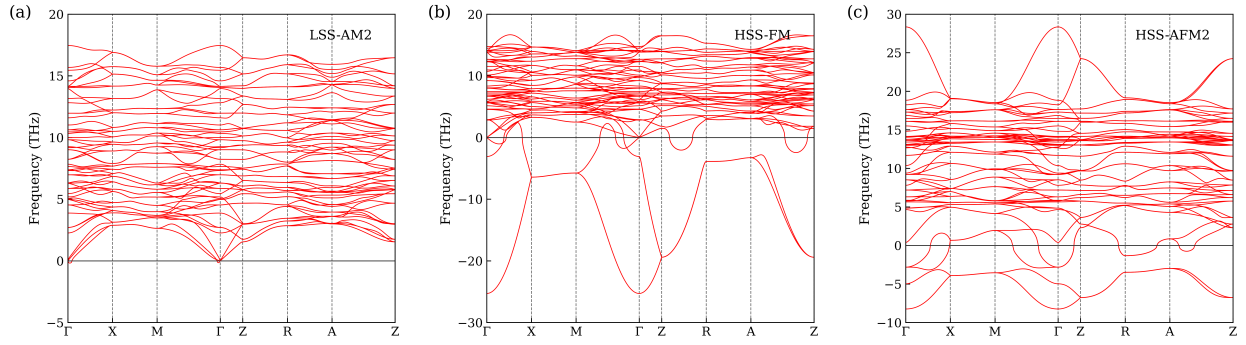


FIG. S3. Phonon spectra of LiV_2F_6 for different structures and magnetic orders.