Supplementary material: Study on lattice dynamics and thermal conductivity of fluorite AF_2 (A = Ca, Sr, Ba) based on first principles

Peipei Liu,[1] Yinchang Zhao,[1,*] Xichang Wang,[1] Jun Ni,[2,3] and Zhenhong Dai[1,†]

[1]*Department of Physics, Yantai University, Yantai 264005, People's Republic of China*

[2]*State Key Laboratory of Low-Dimensional Quantum Physics, Department of Physics, Tsinghua University, Beijing 100084, People's Republic of China*

[3]*Frontier Science Center for Quantum Information, Beijing 100084, People's Republic of China*

(Dated: March 5, 2024)

PACS numbers: 65.40.-b, 66.70.-f, 63.20.-e, 72.20.-i

Keywords: fluorite structure, first-principles calculation, thermal transport properties, lattice thermal conductivity

A. Supplementary Figures.

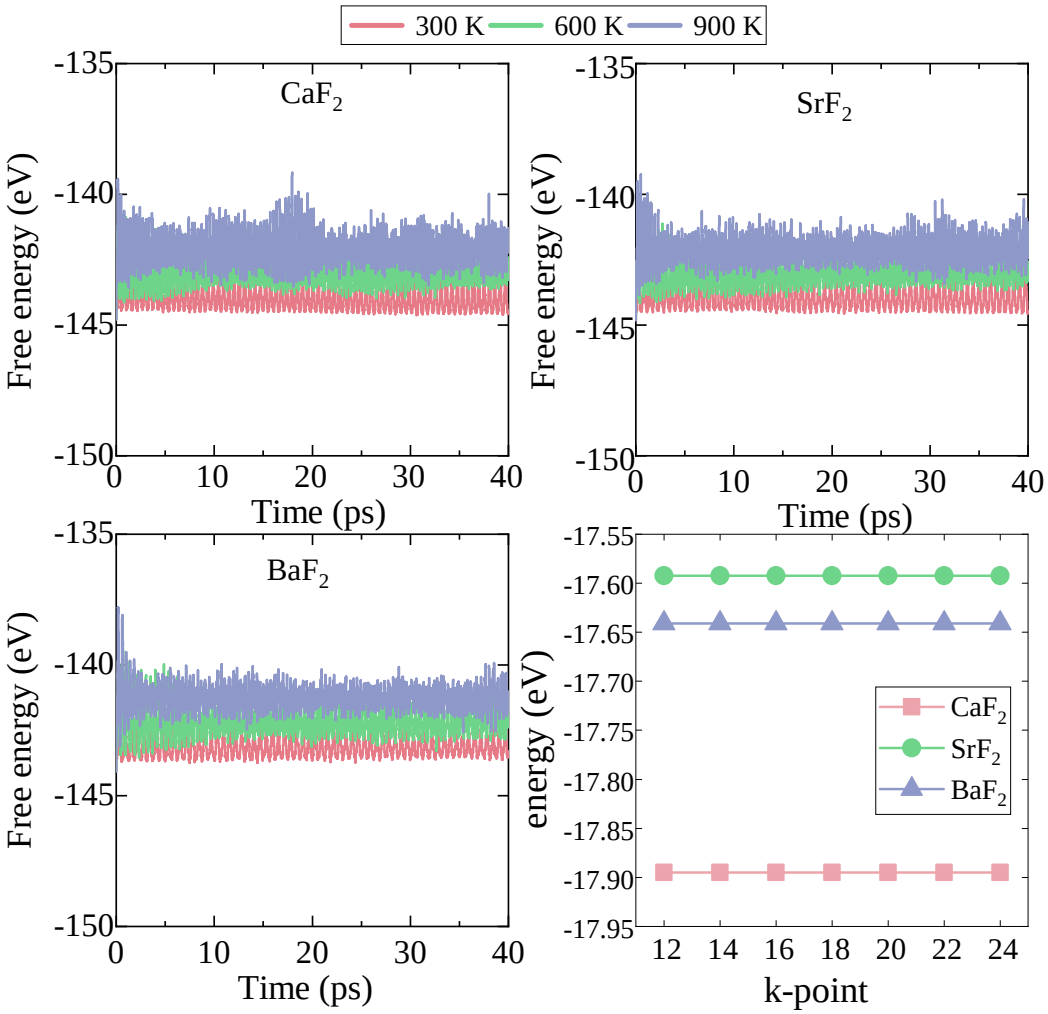

Fig. S1. (Color online). The free energy as the function of time for AF_2 (A = Ca, Sr, Ba) at 300 K, 600 K and 900 K.

S1 are the results of AIMD simulations for the CaF_2, SrF_2 and BaF_2. It is no significant changes in free energy for these three materials, which exhibit these three materials are stable.

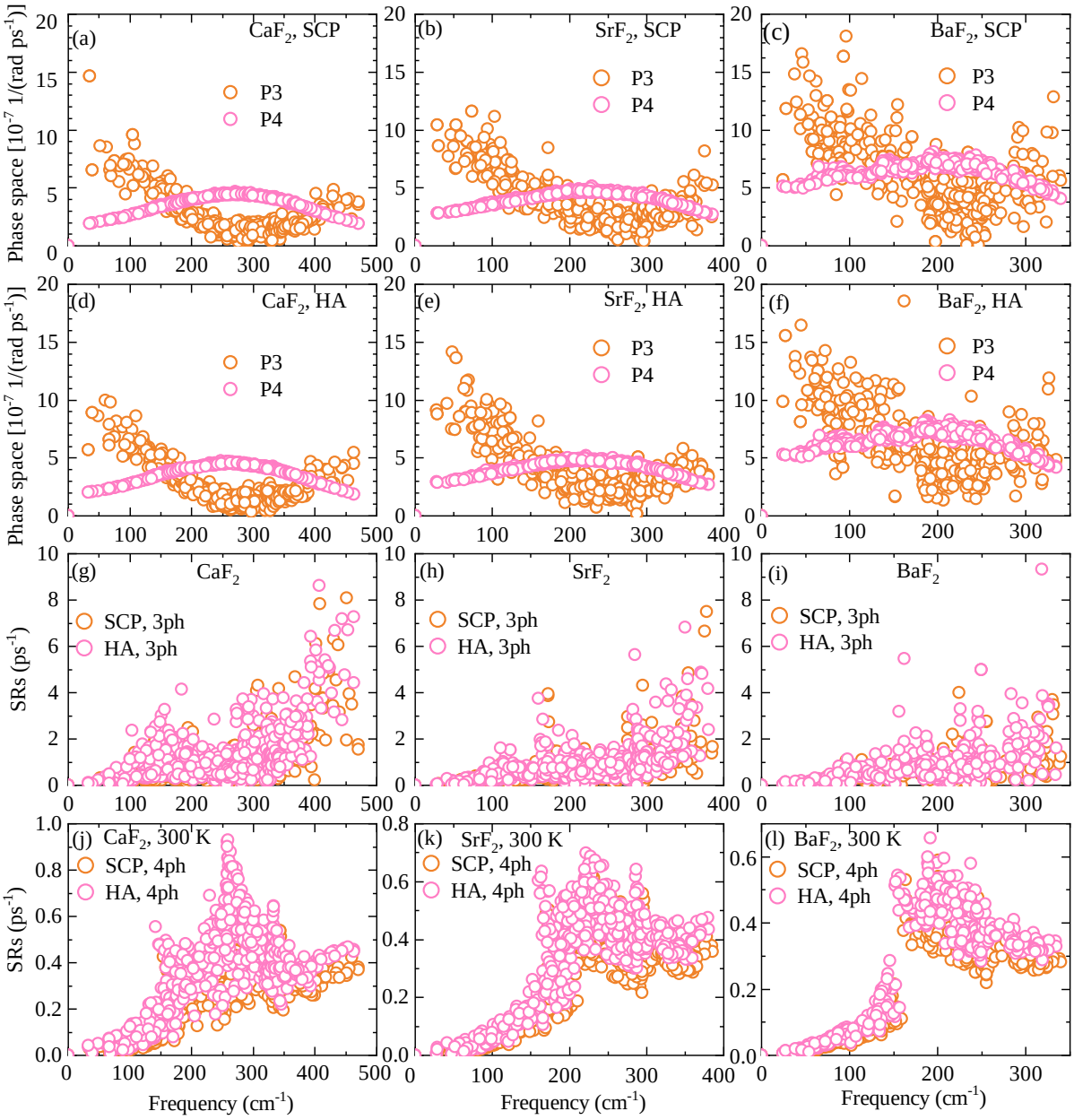

Fig. S2. (Color online). The scattering phase space of (a) CaF_2, (b) SrF_2 and (c) BaF_2 at 300 K was calculated by SCP approximation. In addition, we calculated the 3ph SRs and 4ph SRs of CaF_2, SrF_2 and BaF_2 at 300 K using SCP approximation and HA approximation, respectively.

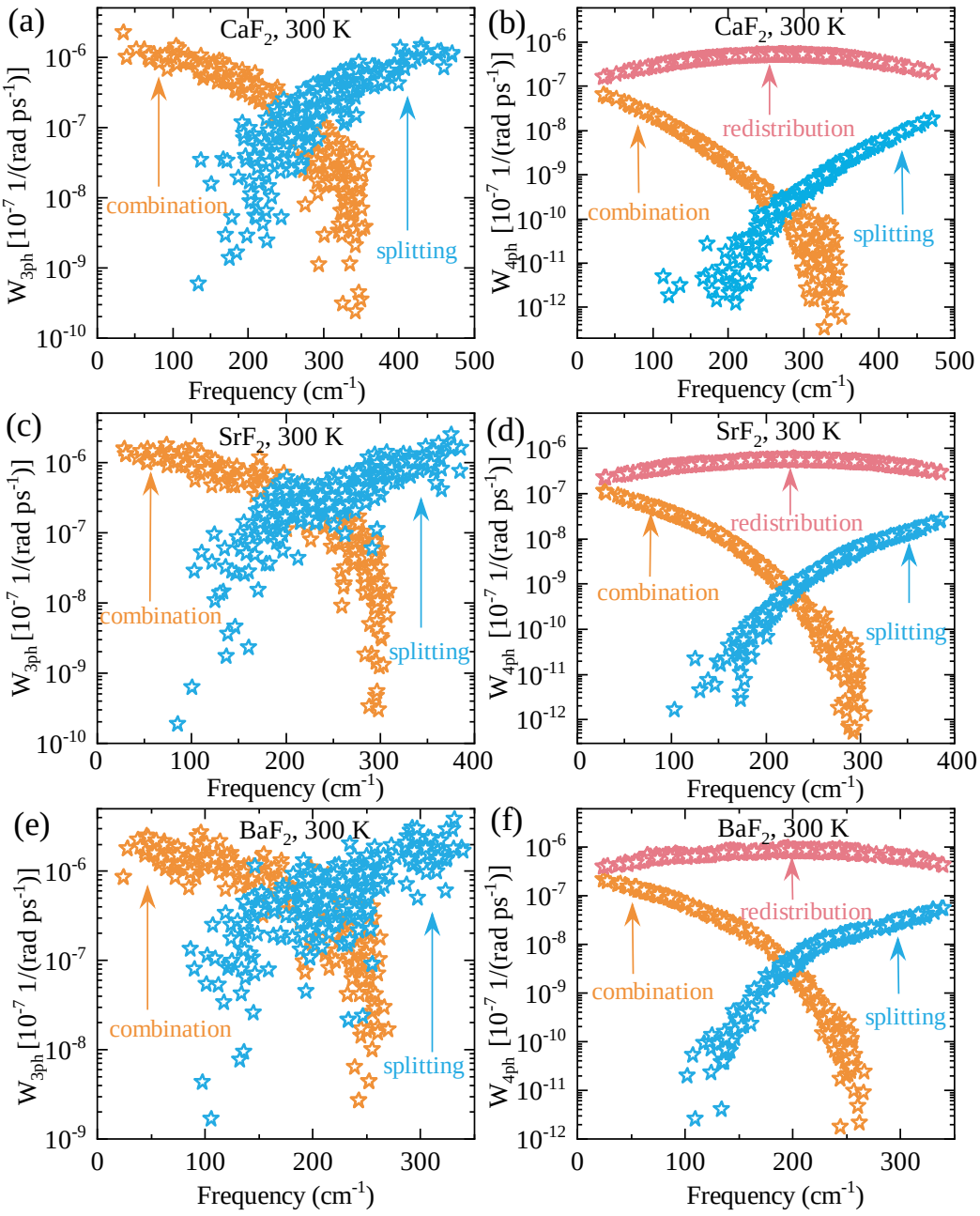

Fig. S3. (Color online). Decomposed 3ph scattering phase spaces into the splitting ($\lambda \rightarrow \lambda^{'} + \lambda^{''}$) and combination ($\lambda + \lambda^{'} \rightarrow \lambda^{''}$) processes at 300 K for (a) CaF_2, (c) SrF_2 and (e) BaF_2. Decomposed 4ph scattering phase spaces into the splitting ($\lambda \rightarrow \lambda^{'} + \lambda^{''} + \lambda^{'''}$), redistribution ($\lambda + \lambda^{'} \rightarrow \lambda^{''} + \lambda^{'''}$) and recombination ($\lambda + \lambda^{'} + \lambda^{''} \rightarrow \lambda^{'''}$) processes at 300 K for (b) CaF_2, (d) SrF_2 and (f) BaF_2. All of the above results were obtained using SCP method.

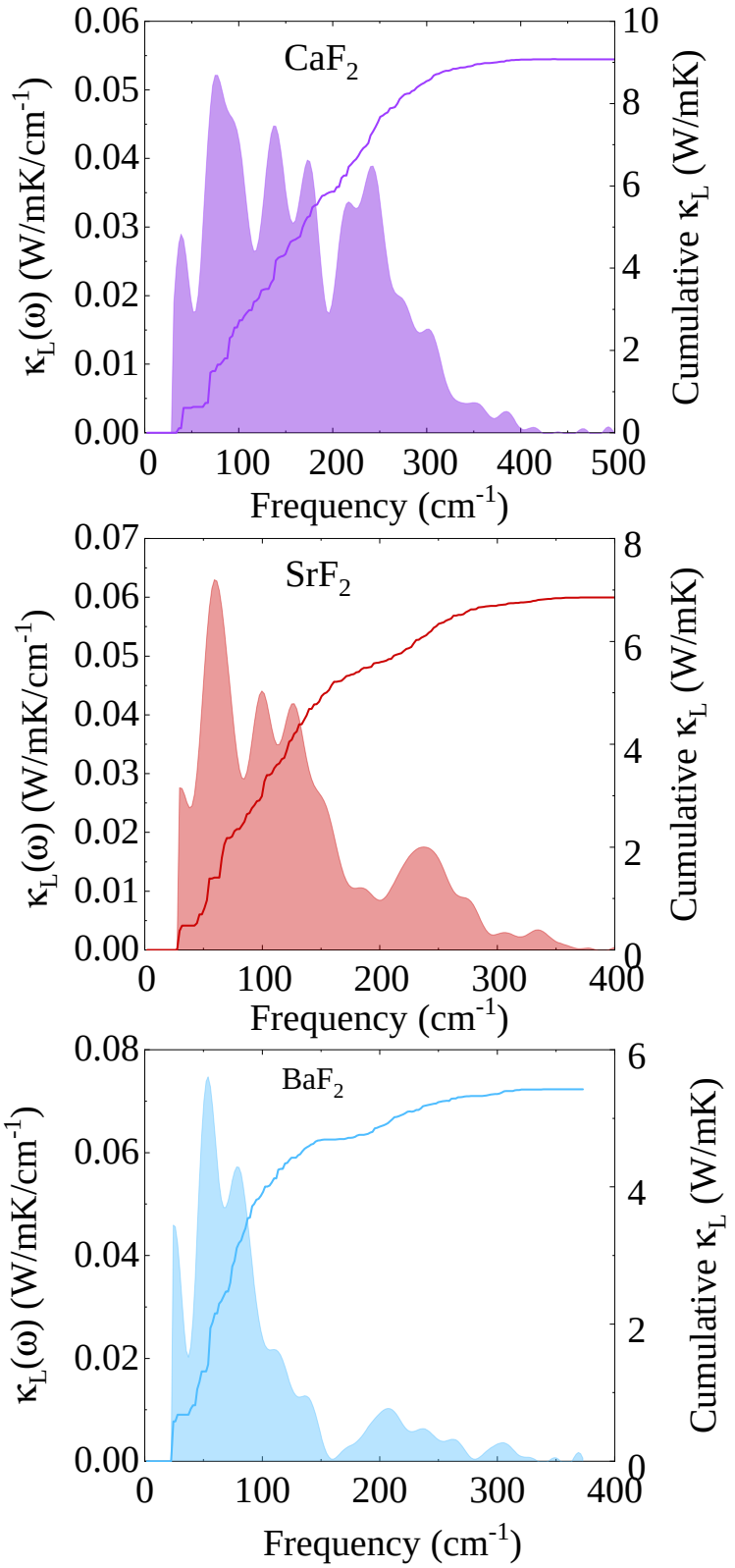

Fig. S4. (Color online). Lattice thermal conductivity spectrums κ_L (ω) (the filled section below the curves) and relevant cumulative κ_L at 300 K for AF_2 (A = Ca, Sr, Ba).

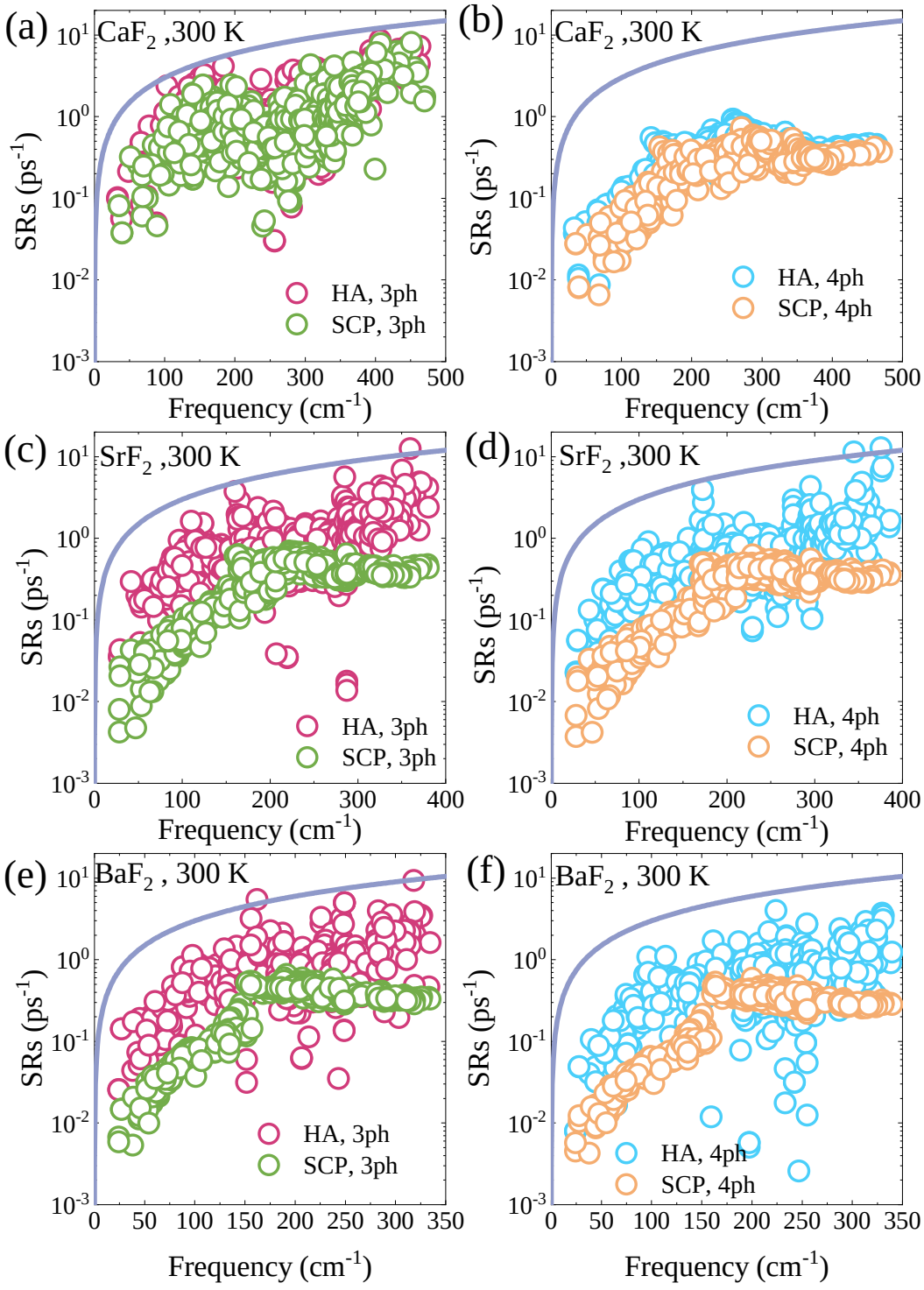

Fig. S5. (Color online). The calculated 3ph SRs and 4ph SRs for AF_2 (A = Ca, Sr, Ba) computed in HA approximations and SCP approximations.

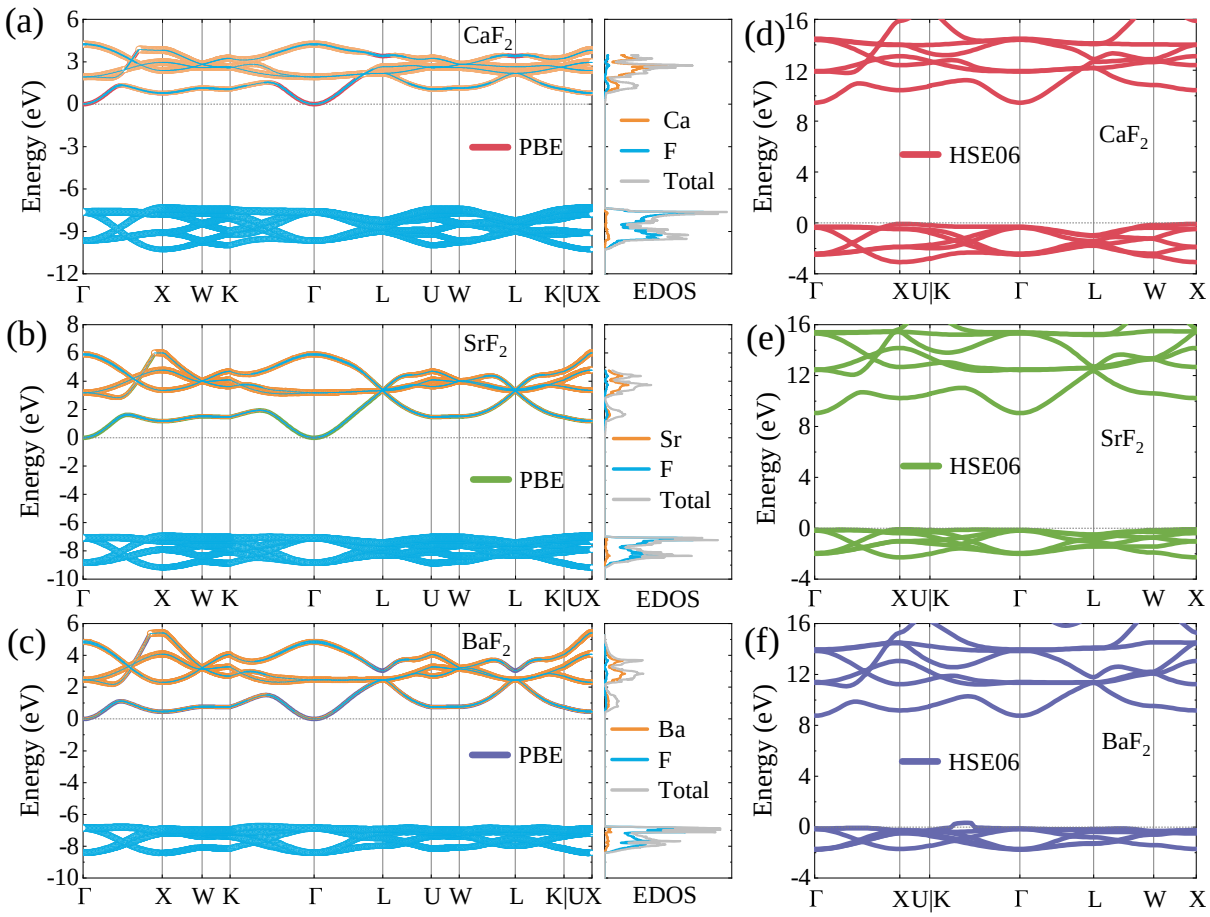

Fig. S6. (Color online). (a)-(c) The band structure of AF_2 (A = Ca, Sr, Ba) and the corresponding density of electronic states (EDOS) are calculated by using PBE functional. (d)-(f) The band structure calculated by HSE06 functional.

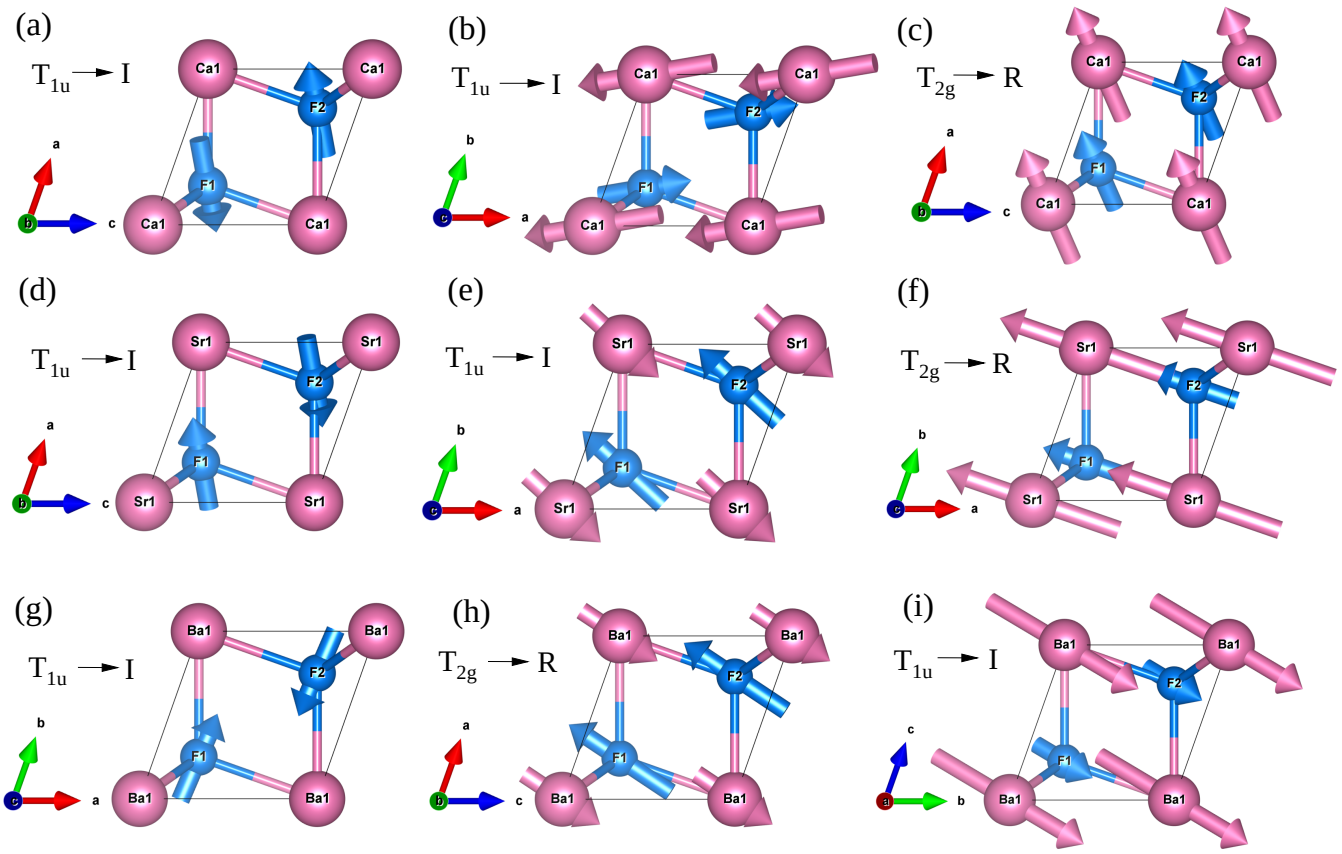

Fig. S7. (Color online). (a)-(i) are the vibration modes of the 1st phonon branch, the 4th phonon branch and the 7th phonon branch of each atom for AF_2 (A = Ca, Sr, Ba) at the Γ point and the Raman modes of the phonon branch.

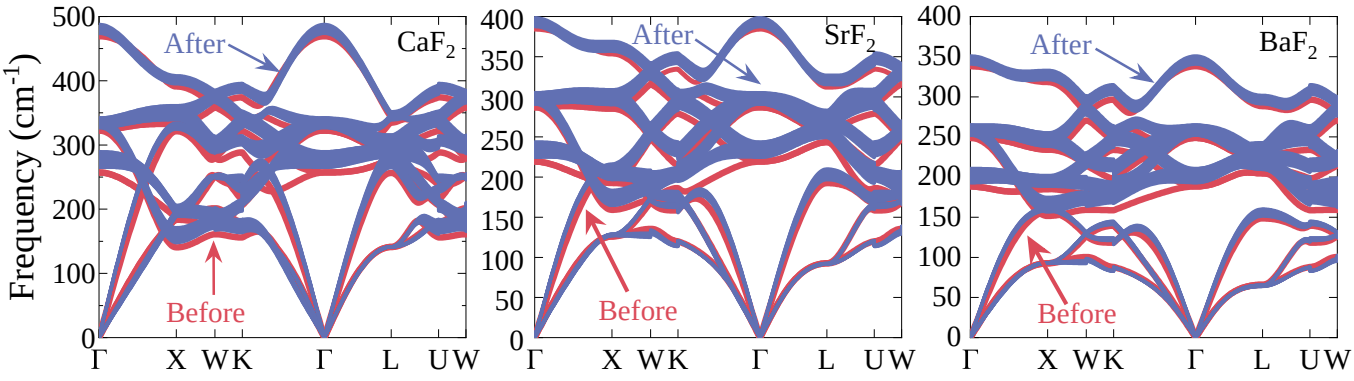

Fig. S8. (Color online). The phonon spectrum of AF_2 (A = Ca, Sr, Ba) before and after anharmonic correction.

* y.zhao@ytu.edu.cn

† zhdai@ytu.edu.cn