Supporting Information

Synthesis, crystal and electronic structures, nonlinear optical properties, and magnetic properties of two thiophosphates: $KInP_2S_7$ and $KCrP_2S_7$.

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1. **Figure S1**. Powder X-ray diffraction results of $KInP_2S_7$.
2. **Figure S2.** 3D isosurfaces of the electron localization function (ELF) for $KInP_2S_7$ with η=0.75, K: green, In: pink, P: black, S: yellow.
3. **Figure S3.** Kubelka-Munk diffuse reflectance spectrum of $KInP_2S_7$.
4. **Figure S4**. Tauc plots for allowed direct and indirect transitions of $KInP_2S_7$.
5. **Figure S5**. SHG intensities of $KInP_2S_7$ and $AgGaS_2$ @2.09 μm measured at variable particle size samples.
6. **Table S1**. LDT measurement results of $KInP_2S_7$ and $AgGaS_2$ @ 1064 nm measured at samples of particle size of 225 μm.

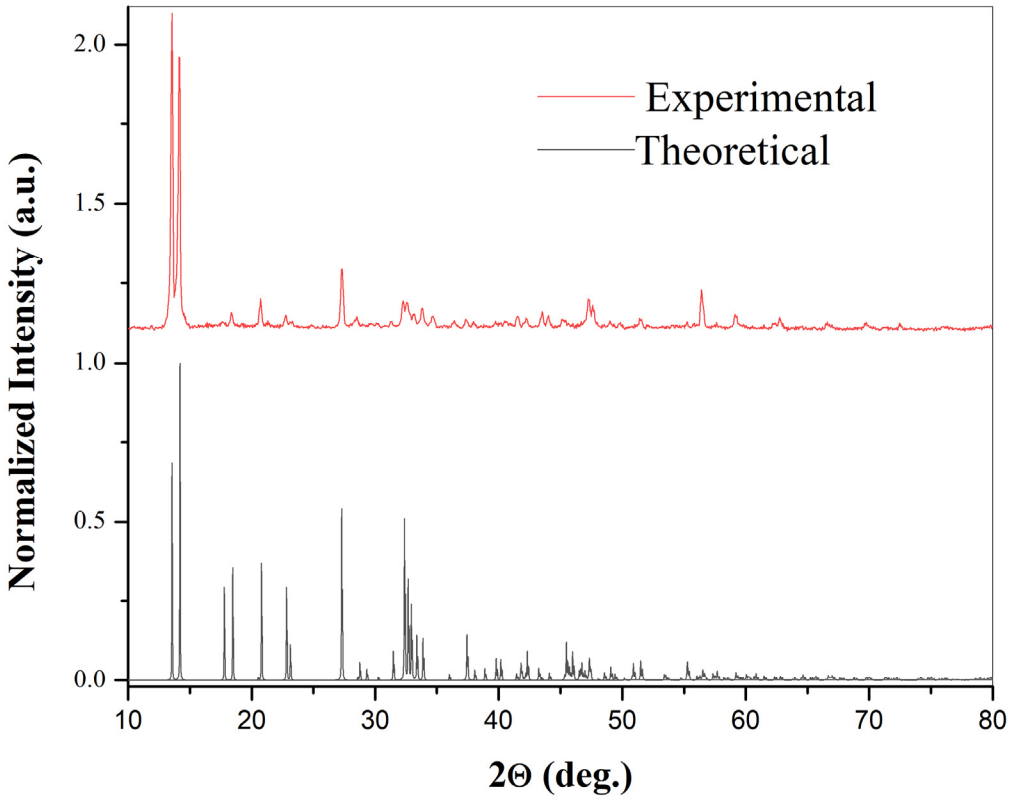

Figure S1. Powder X-ray diffraction results of $KInP_2S_7$.

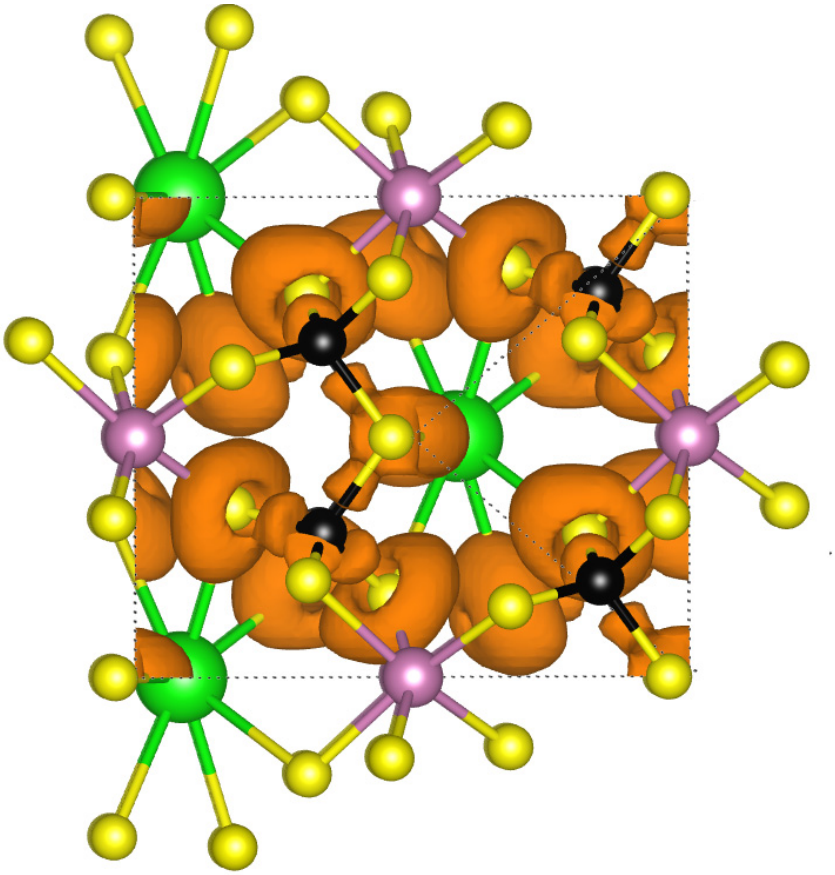

Figure S2. 3D isosurfaces of the electron localization function (ELF) for $KInP_2S_7$ with η=0.75, K: green, In: pink, P: black, S: yellow.

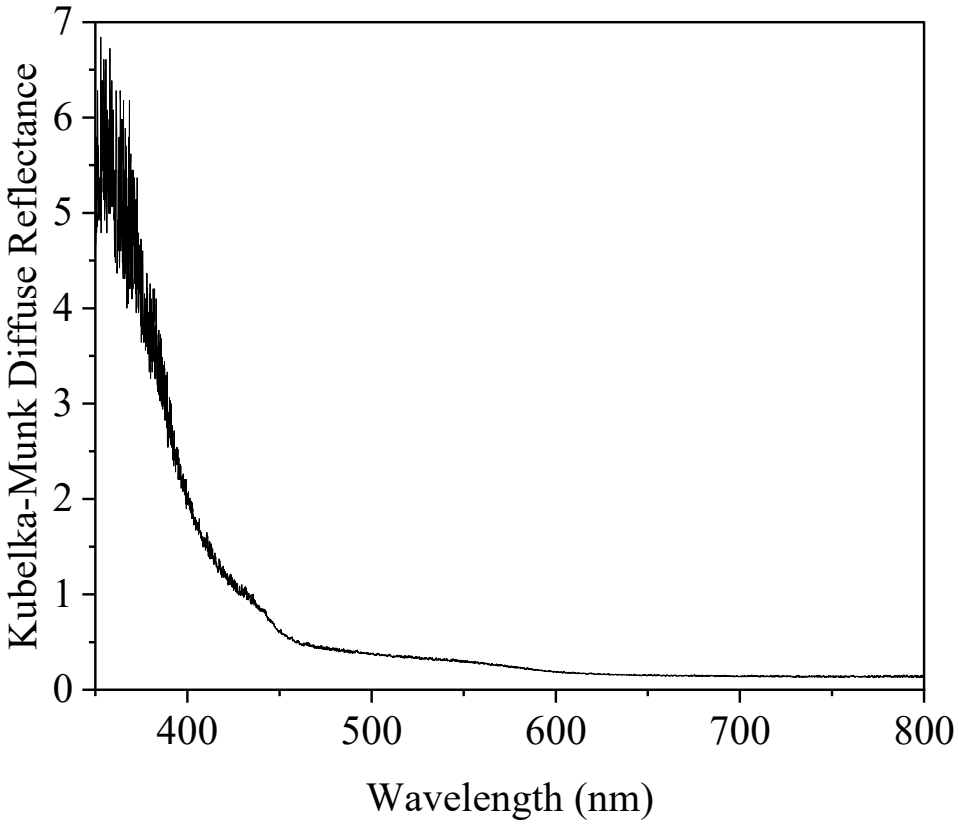

Figure S3. Kubelka-Munk diffuse reflectance spectrum of $KInP_2S_7$.

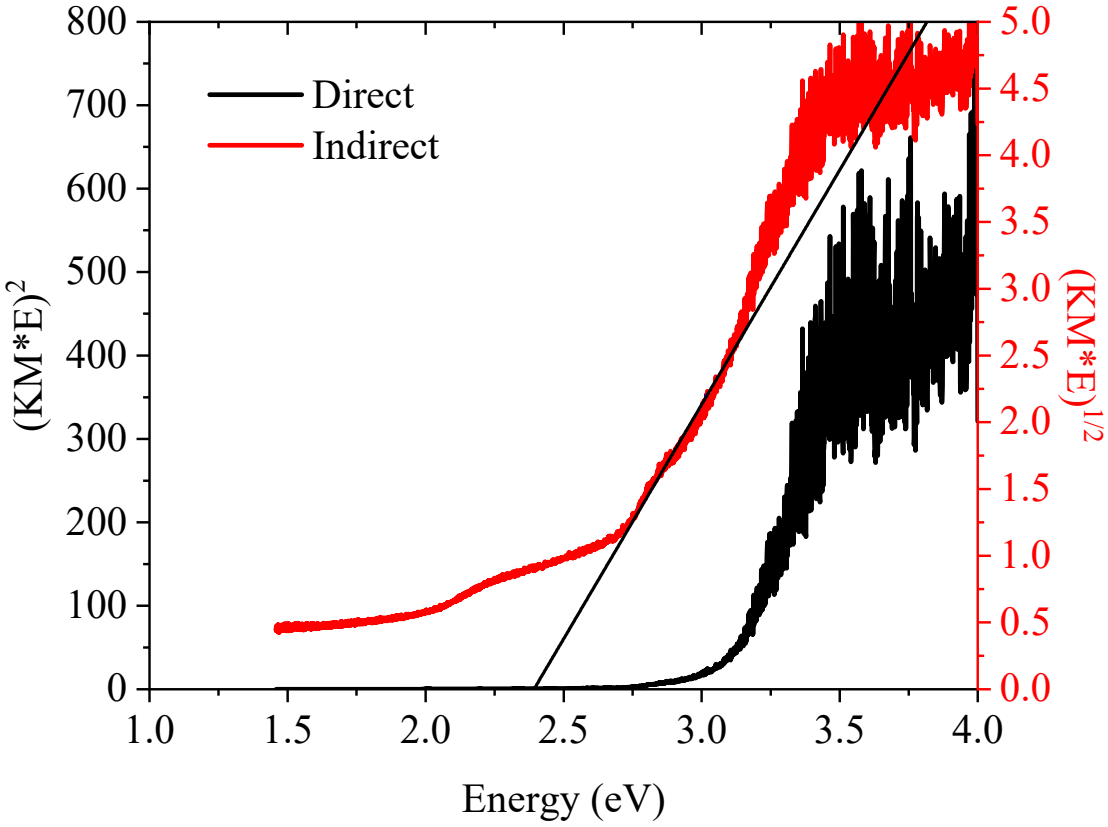

Figure S4. Tauc plots for allowed direct and indirect transitions of $KInP_2S_7$.

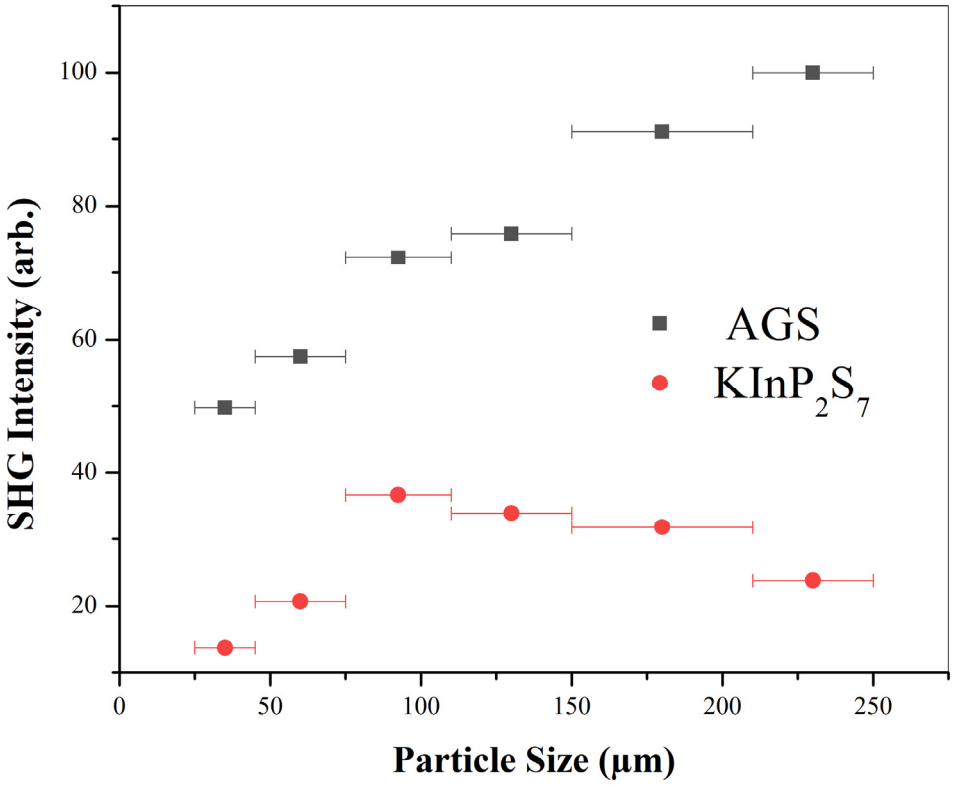

Figure S5. SHG intensities of $KInP_2S_7$ and $AgGaS_2$ @2.09 μm measured at variable particle size samples.

Table S1. LDT measurement results of $KInP_2S_7$ and $AgGaS_2$ @ 1064 nm measured at samples of particle size of 225 μm.

	damage energy (mJ)	**spot diameter (mm)**	**LDT (MW/cm2)**	**LDT (× AGS)**
$AgGaS_2$	0.58	0.5	29.6	1
$KInP_2S_7$	3.02	0.5	154	5.2