Supplementary Information

Phonon spectrum of $Pr_2Zr_2O_7$ and $Pr_2Ir_2O_7$ as an evidence of coupling of the lattice with electronic and magnetic degrees of freedom.

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I. LINE WIDTH TEMPERATURE DEPENDENCE

$T_{2g}^{(1)}$, $T_{2g}^{(2)}$, and E_g modes in $Pr_2Zr_2O_7$ and $T_{2g}^{(2)}$ modes in $Pr_2Ir_2O_7$ were fit by the extended Klemens' model, which takes the form of,

$$\Gamma(T) = \Gamma_0 + A(1 + 2n_B(\omega/2, T)) + B(1 + 3n_{\mathrm{B}}(\omega/3, T) + 3(n_B(\omega/3, T))^2). \quad (1)$$

Mode	Γ_0 (cm^{-1})	A (cm^{-1})	B (cm^{-1})
$T_{2g}^{(1)}$	1.901	4.811	0.188
E_g	4.239	6.861	0.048
A_{1g}	0	7.571	0.232

TABLE S1: Results of fitting for $Pr_2Zr_2O_7$.

$T_{2g}^{(1)}$, E_g, $T_{2g}^{(2)}$, and A_{1g} modes in $Pr_2Ir_2O_7$ was fit by the model of decay into electron-hole pairs, which takes the form of,

$$\Gamma(T) = \Gamma_0 + F(n_{\mathrm{F}}(\hbar\omega_a, T) - n_{\mathrm{F}}(\hbar\omega_a + \hbar\omega_{\mathrm{ph}}, T)). \quad (2)$$

Mode	Γ_0 (cm^{-1})	F (cm^{-1})	ω_a (cm^{-1})
$T_{2g}^{(1)}$	13.017	66.444	243.654
E_g	38.430	619.369	775.777
$T_{2g}^{(2)}$	14.585	40.0453	256.860
A_{1g}	7.230	30.701	275.574

TABLE S2: Results of fitting for $Pr_2Ir_2O_7$.

II. ROOM TEMPERATURE FITTING

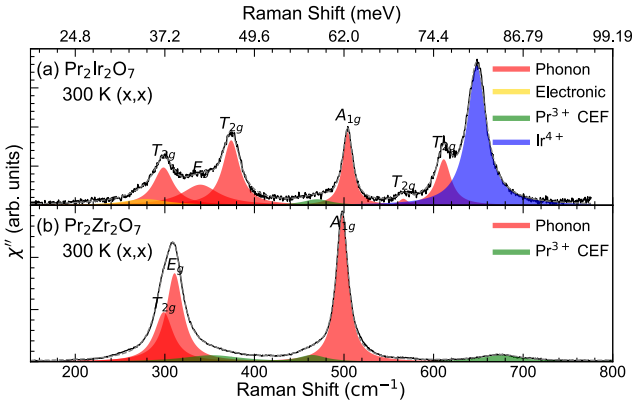

FIG. S1: Raman scattering spectra of (a) $Pr_2Ir_2O_7$ and (b) $Pr_2Zr_2O_7$ at 300 K in parallel polarization configuration. Solid black curve shows the experimental data after a linear background subtraction. Dashed grey line shows the overall fitting.

III. TEMPERATURE DEPENDENCE OF THE PHONON INTENSITY

We estimate the temperature dependence of the intensity of phonons as their spectral weight $\int \chi''(\omega)\delta\omega$, which should stay constant with temperature. Temperature dependence of the phonon spectral weight is shown in Fig. S2. While the intensities measured in Raman scattering are not absolute, but arbitrary units, we still can see that the spectral weight of the $Pr_2Zr_2O_7$ phonons is two orders of magnitude higher than that of $Pr_2Ir_2O_7$, due to the metallic properties of the latter material.

For $Pr_2Zr_2O_7$ spectra, all phonons exhibit a constant spectral weight with temperature, except for $T_{2g}^{(2)}$ phonon which is mixed with the crystal field excitation at high temperature. $T_{2g}^{(2)}$ is shown to be interacting with $A_{1g} \rightarrow E_g$ transition between crystal field levels. Spectral weight will be defined by these interactions, however, the theory which describes a general Fano approach to such interactions does not describe the intensities, which will depend on the particular type of interactions.

The low intensity of the $Pr_2Ir_2O_7$ spectra makes spectral weight of the $Pr_2Ir_2O_7$ phonons susceptible to a systematic error due to small mechanical perturbations of the measuring system on cooling, keeping the intensity of the spectra consistent is experimentally challenging. The decrease of the spectral weight of T_{2g} and E_g phonons on cooling is very small and cannot be distinguished from that introduced by a micron-sized mechanical shift. The change of the intensity of the A_{1g} follows roughly the resistivity of $Pr_2Ir_2O_7$ [1]. However, from our point of view, this information does not provide enough novelty to include it in the main text of the paper.

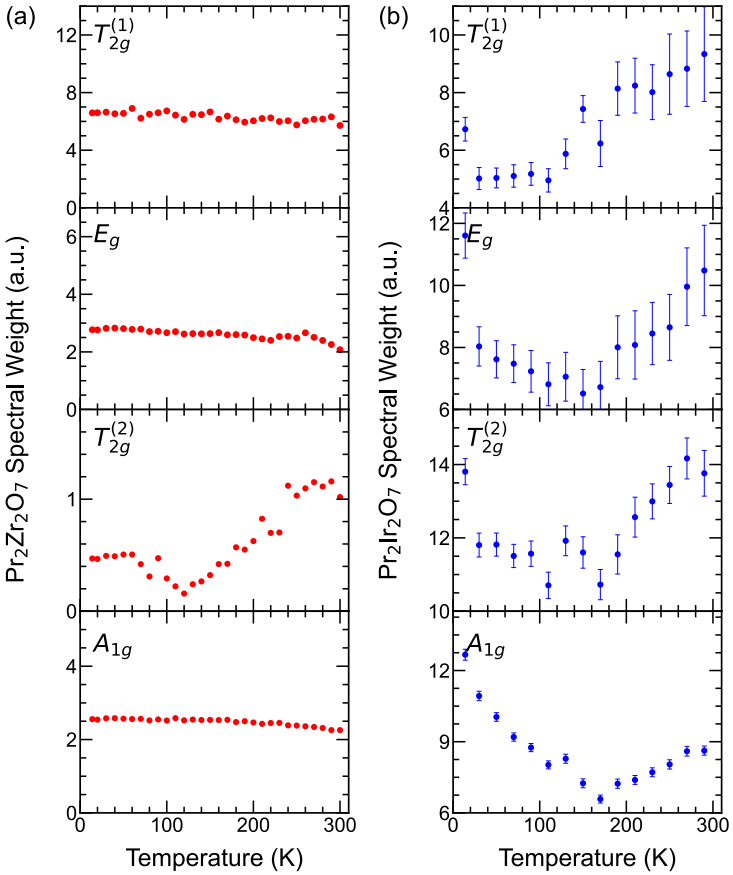

FIG. S2: Temperature dependence of the phonon intensity in (a) $Pr_2Zr_2O_7$ and (b) $Pr_2Ir_2O_7$.

IV. POLARIZATION DEPENDENCE

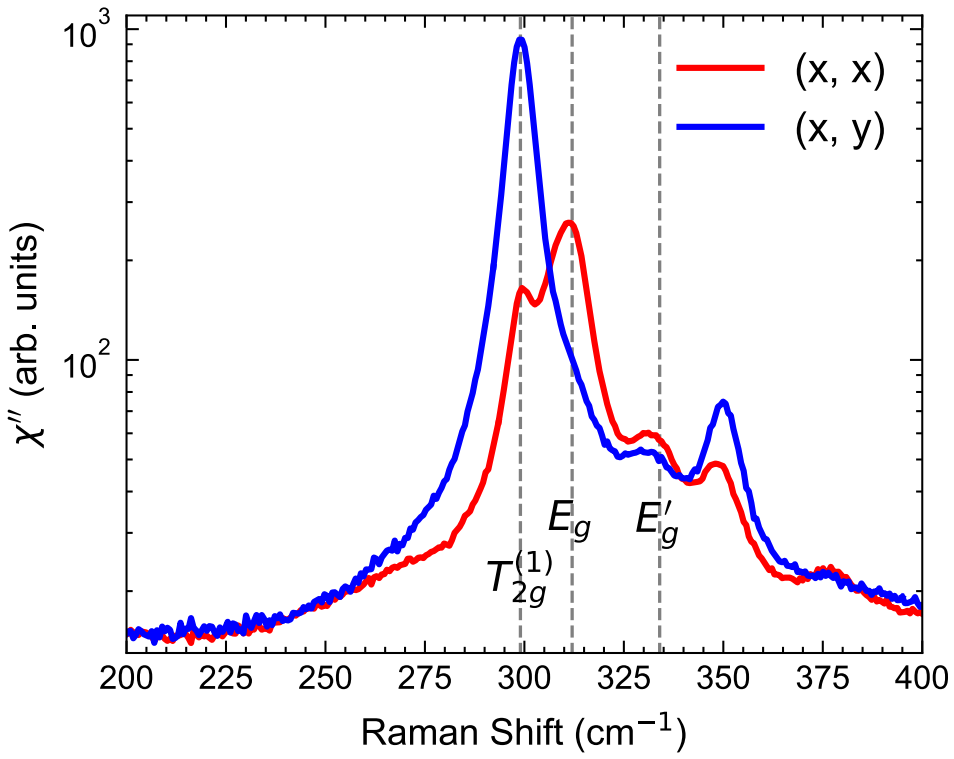

FIG. S3: Polarized Raman scattering spectra measured from (100) facet at 14 K. Polarization dependence reveals a difference between E_g and T_{2g}.

V. ELECTRONIC BAND STRUCTURE

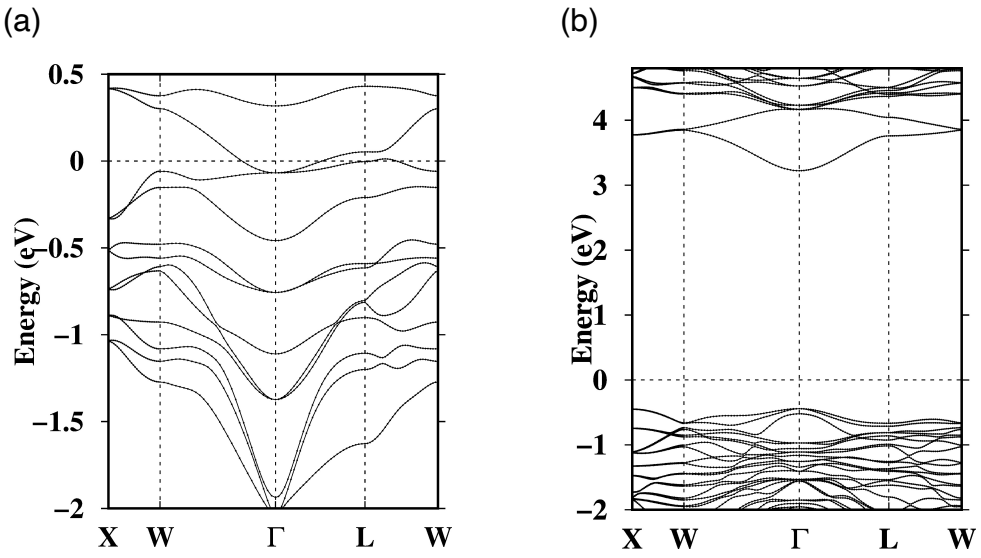

FIG. S4: The nonmagnetic band structure calculated under PBE+SOC for (a) $Pr_2Ir_2O_7$ and (b) $Pr_2Zr_2O_7$. $Pr_2Ir_2O_7$ shows a band touching slightly below the Fermi level, while $Pr_2Zr_2O_7$ shows an insulating gap $E_{\rm gap} = 3.66$ eV.

[1] S. Nakatsuji, Y. Machida, Y. Maeno, T. Tayama, T. Sakakibara, J. van Duijn, L. Balicas, J. N. Millican, R. T. Macaluso, and Julia Y. Chan. Metallic spin-liquid behavior of the geometrically frustrated kondo lattice $pr_2ir_2o_7$. *Phys. Rev. Lett.*, 96:087204, Mar 2006.