

# Supplemental Material for: Controlling the magnetic state of the proximate quantum spin liquid $\alpha$ -RuCl<sub>3</sub> with an optical cavity

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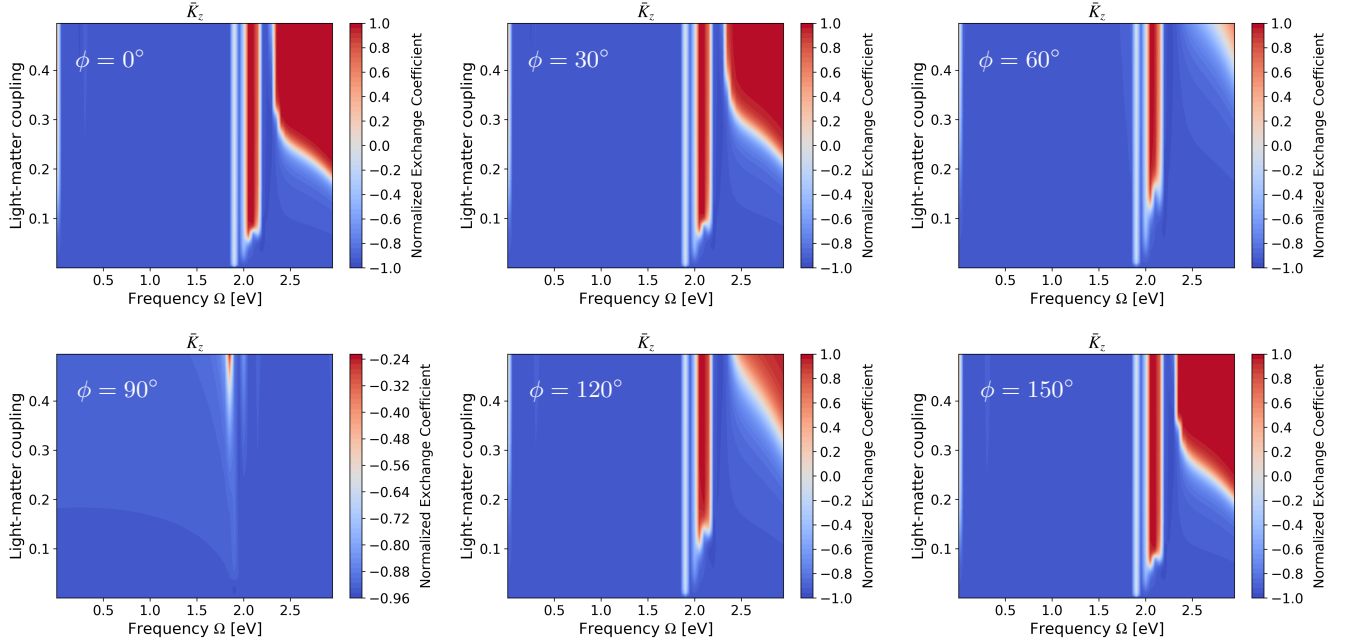
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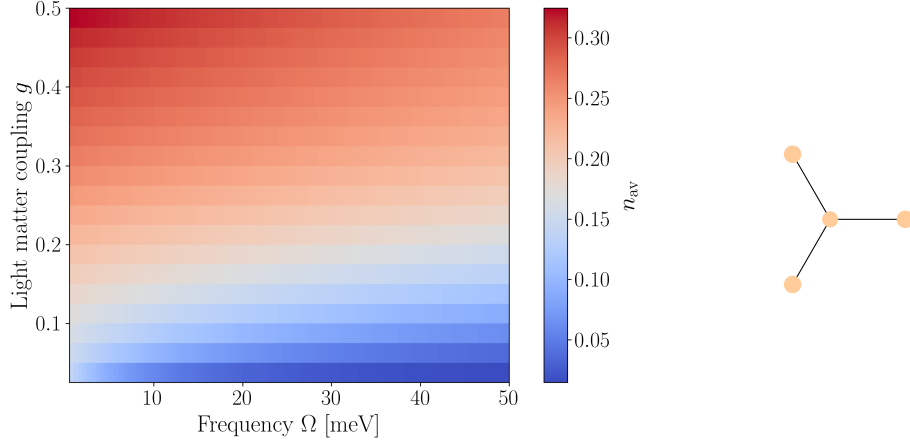
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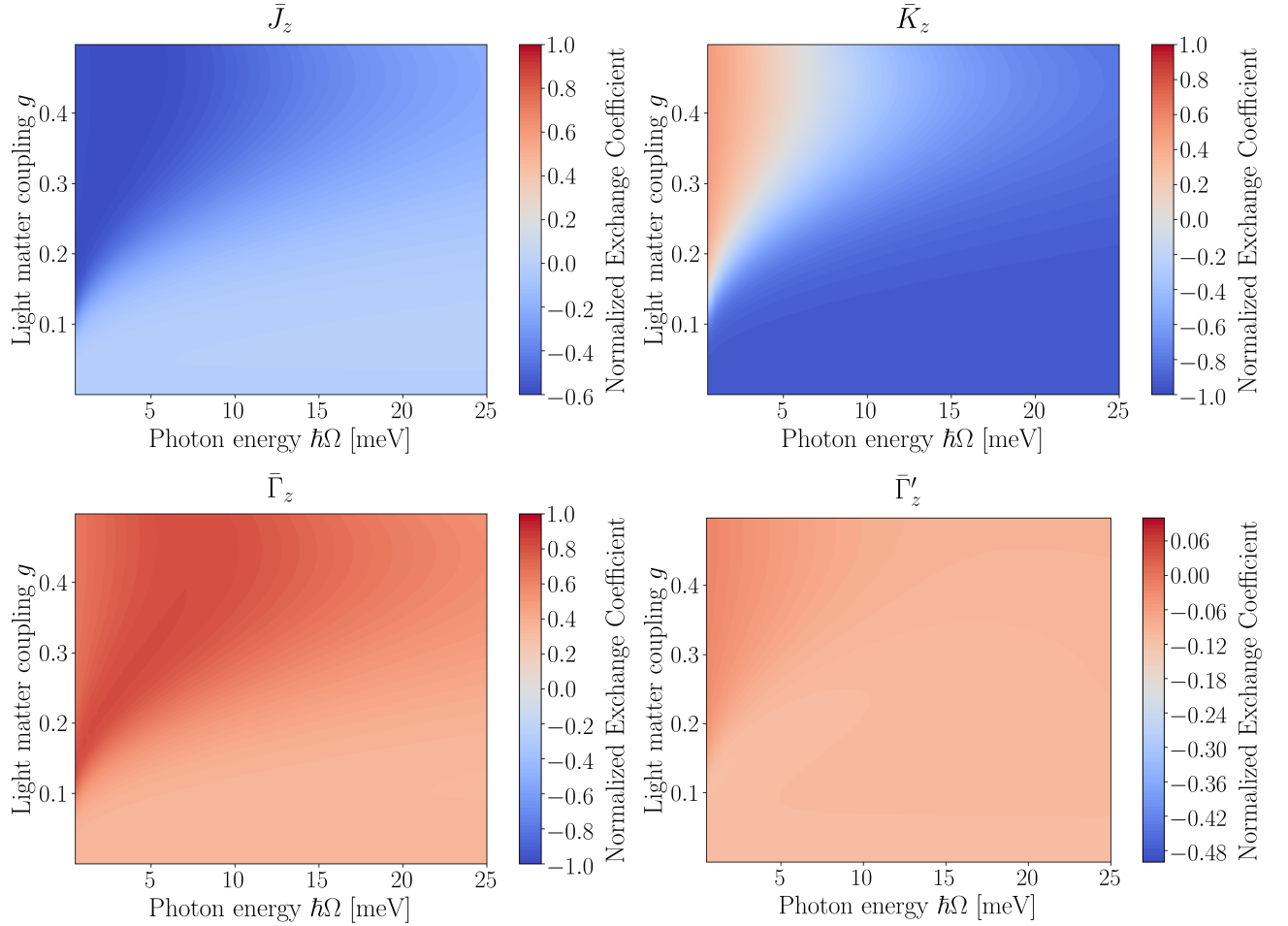
Supplementary Figure 1. **Polarization dependence of the Kitaev interaction for a linearly polarized cavity mode.** Effective Kitaev interaction  $\bar{K}$  as a function of the effective light-matter coupling  $\bar{g} = 2g\sqrt{n_{\text{av}}}$  and frequency  $\Omega$  for a cavity with  $n_{\text{av}} = 1$ . The different panels correspond to polarization vectors making an angle  $\phi$  with the  $x$ -axis. Due to the  $C_3$  symmetry of the magnetic system, the polarization dependence shows a period of  $\phi_0 = 2\pi/3$ .

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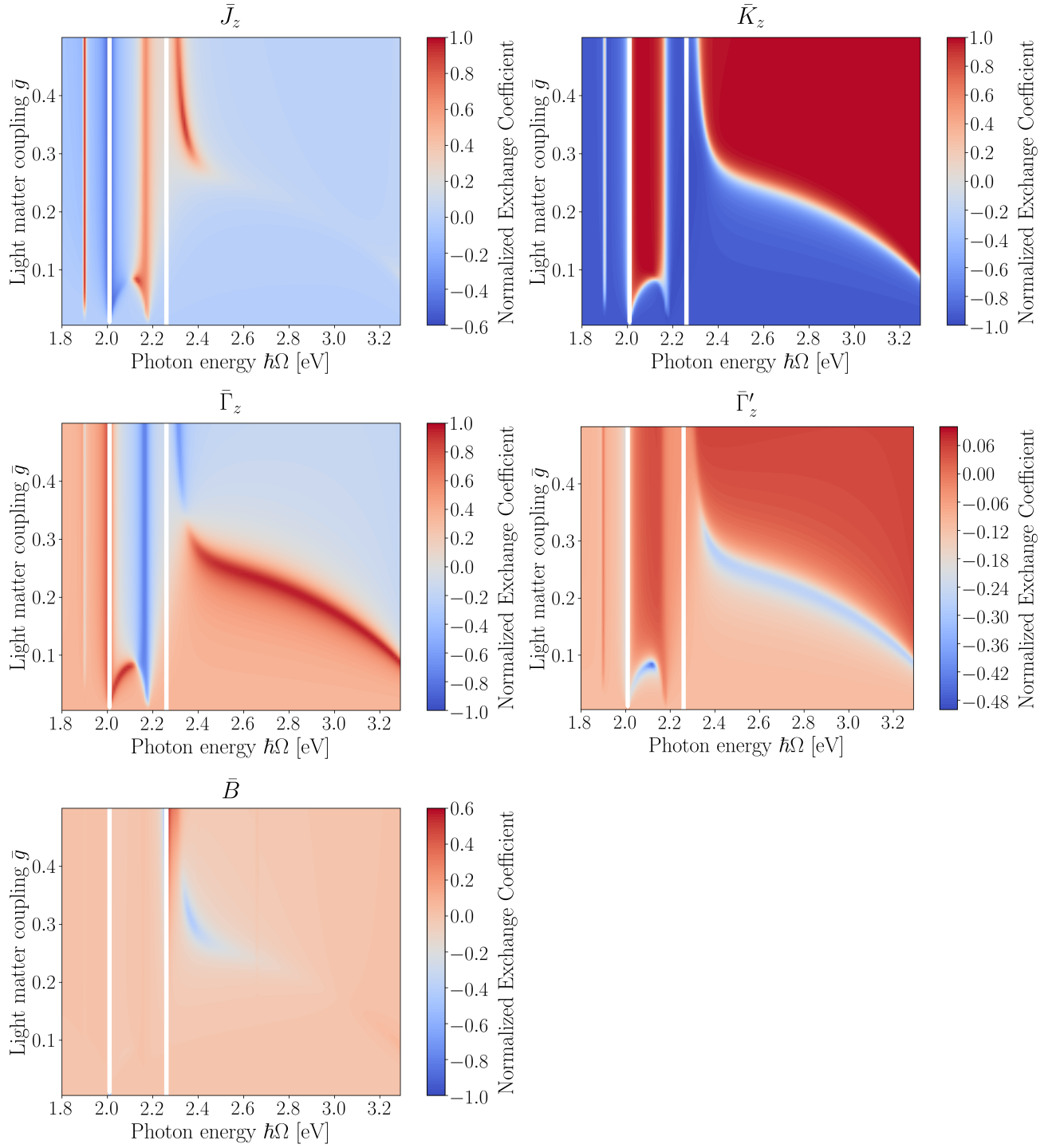
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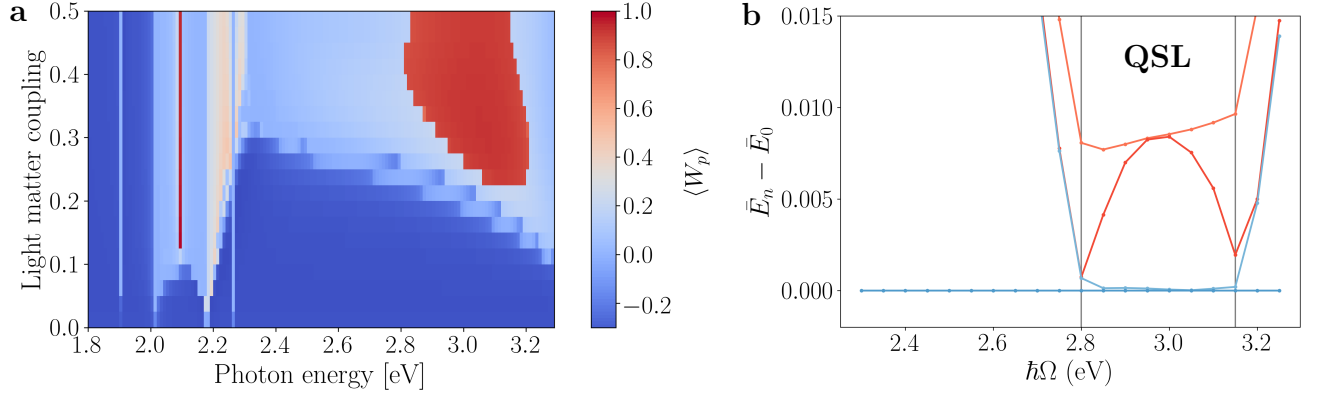
Supplementary Figure 2. **Photon occupation of an electronic cluster.** Photon occupation of the ground state of a four site Ru cluster described by the Hamiltonian in Eqs. 3 and 15 of the main text. The Ru cluster is schematically shown to the right.



Supplementary Figure 3. **Spin parameters of the dark cavity.** Normalized magnetic exchange interaction  $\bar{J}$ , Kitaev interaction  $\bar{K}$ , and anisotropy interactions  $\bar{\Gamma}$  and  $\bar{\Gamma}'$  as a function of the light-matter coupling  $g$  and the photon energy  $\hbar\Omega$  in the zero photon sector  $n_{\text{av}} = 0$ .



Supplementary Figure 4. **Spin parameters of the seeded cavity.** Normalized magnetic exchange interaction  $\bar{J}$ , Kitaev interaction  $\bar{K}$ , anisotropy interactions  $\bar{\Gamma}$  and  $\bar{\Gamma}'$  and induced magnetic field  $\bar{B}$  as a function of the effective light-matter coupling  $\bar{g}$  and the photon energy  $\hbar\Omega$  in the zero photon sector  $n_{\text{av}} = 1$ .



Supplementary Figure 5. **Validation of the Kitaev quantum spin liquid state.** **a**, Expectation value of the flux operator  $W_p$  as a function of photon energy  $\hbar\Omega$  and light-matter coupling  $\bar{g}_{\text{eff}} = 2g_{\text{eff}}\sqrt{n_{\text{av}}}$  for fixed average photon number  $n_{\text{av}} = 1$ . **b**, Lowest energy eigenstates of the effective photo-renormalized spin Hamiltonian as a function of photon energy  $\hbar\Omega$  for  $\bar{g}_{\text{eff}} = 0.4$  (corresponding to a horizontal slice through panel **a**). The blue lines show the energies of the ground state and the first excited state, while the red lines show the energies of the second and third excited state. For  $\hbar\Omega = 2.8 - 3.2$  eV an excitation gap opens and a topological ground state degeneracy appears, indicating the appearance of a gapped Kitaev quantum spin liquid state.