

Inducing a quantum spin liquid state in the approximate Kitaev material α -RuCl₃

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One way to characterize quantum spin-liquids is through the fractionalization of spin excitations. A prime example of this is found in the exactly solvable Kitaev model of spin-1/2 moments with anisotropic exchange interactions on a tri-coordinated lattice [1]. To find examples of this kind of physics in nature turns out to be challenging. The currently best known examples of materials in which Kitaev-like physics plays a central role are the layered spin-orbit entangled $J=1/2$ systems Na₂IrO₃, α -Li₂IrO₃, and α -RuCl₃. However, these materials all possess additional interactions, which, among other, lead to a magnetically ordered state at low temperature preventing the formation of a pure Kitaev spin-liquid (KSL) state. Apart from the ongoing quest for materials showing a true KSL ground state, one can also destabilize the magnetic order in the existing materials, which potentially can induce the sought after KSL state. In this contribution I will discuss two methods to destabilize magnetic order in α -RuCl₃. The first one is through the application of an in-plane magnetic field. Though it has been shown by various authors that this indeed leads to suppression of the ordered state in α -RuCl₃, the nature of the field-induced state is not fully clear. The most intriguing suggestion comes from heat transport experiments showing the existence of a quantized thermal Hall conductivity [2] just above the critical field, which is a Hallmark of a KSL. Our spectroscopic Raman and THz experiments do not show evidence for a field induced Kitaev spin-liquid state, but rather, combined with exact diagonalization results, show the existence of a quantum disordered partially aligned high field phase characterized by a gapped continuum of magnetic excitations combined with single particle and bound state resonances. The second approach is a pump-probe method which creates holon and doublon excitations. These excitations are found to couple efficiently to magnetic excitations which in turn disorder the magnetically ordered state. For sufficiently high excitation densities the magnetic order is fully suppressed, leading to a quantum disordered magnetic state.

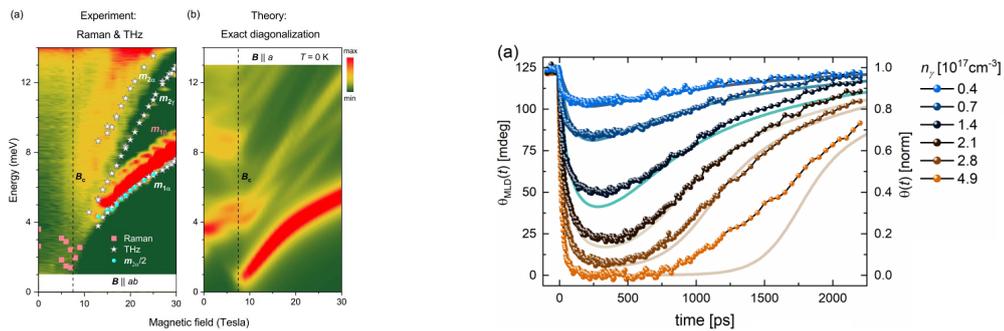


Figure 1. α -RuCl₃ **Left** Field dependent Raman spectra (with superimposed the position of sharp 1 Hz resonances) in false colour representation showing the opening of an excitation gap above 7T and the presence of single particle and bound state resonances. The right panel shows an exact diagonalization calculation of the field dependent Raman result. [3] **Right** pump-probe transients tracking the antiferromagnetic order parameter through magnetic linear dichroism after the creation of doublon-holon excitations. Above a doublon (holon) density of $4.5 \times 10^{17} \text{ cm}^{-3}$ the antiferromagnetic order is fully suppressed for a few hundred ps [4].

References

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- [4] R.B. Versteeg, et al., arXiv:2005.14189; J. Wagner, et al., *Faraday disc.* (2022)